ANALYTICAL INVERSE MODEL FOR POST-EVENT ATTRIBUTION OF PLUTONIUM

A Thesis

by

JAMES CHRISTOPHER MILLER

Submitted to the Office of Graduate Studies of
Texas A&M University
in partial fulfillment of the requirements for the degree of

MASTER OF SCIENCE

December 2008

Major Subject: Nuclear Engineering

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Approved by:

Chair of Committee,	William S. Charlton
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ABSTRACT

Analytical Inverse Model for Post-Event Attribution of Plutonium.

(December 2008)

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Chair of Advisory Committee: Dr. William S. Charlton

An integral part of deterring nuclear terrorism is the swift attribution of any event to a particular state or organization. By quickly being able to identify the responsible party after a nuclear event, appropriate people may be held accountable for their actions. Currently, there is a system in place to determine the origin of nuclear devices and materials from post-event data; however, the system requires significant time to produce an answer within acceptable error margins. Described here is a deterministic approach derived from first principles to solve the inverse problem. The derivation starts with the basic change rate equation and ends in relationships for important nuclear concentrations and device yield. This results in a computationally efficient and timely method for producing an estimate of the material attributes. This estimate can then be used as a starting point for other more detailed methods and reduce the overall computation time of the post-event forensics.

This work focused on a specific type of nuclear event: a plutonium improvised nuclear device (IND) explosion. From post-event isotopic ratios, this method determines the device's pre-event isotopic concentrations of special nuclear material. From the

original isotopic concentrations, the field of possible origins for the nuclear material is narrowed. In this scenario, knowing where the nuclear material did not originate is as important as knowing where it did.

The derived methodology was tested using several cases of interest including simplified and realistic cases. For the simplistic cases, only two isotopes comprised the material being fissioned. In the realistic cases, both Weapons Grade and Reactor Grade plutonium were used to cover the spectrum of possible fissile material to be used by terrorists. The methodology performed very well over the desired energy range. Errors were under two percent from the expected values for all yields under 50 kT. In the realistic cases, competing reactions caused an increase in error; however, these stayed under five percent. As expected, with an increased yield, the error continued to rise, but these errors increased linearly. A sensitivity analysis was performed on the methodology to determine the impact of uncertainty in various physical constants. The result was that the inverse methodology is not overly sensitive to perturbations in these constants.

ACKNOWLEDGEMENTS

I would like to acknowledge Adrienne LaFleur for her valuable insights in discussing the content of this work. I would also like to thank Dr. William Charlton, my committee chair. His guidance and support throughout my first years of graduate school have been instrumental. I would like to thank Dr. Brian Bluhm for his wisdom and insight into my career path and his assistance with obtaining not only summer internships at Los Alamos National Laboratory, but also in applying for doctoral fellowships. Finally, I would like to thank David Grant Ford, Jr. for his assistance in the thesis writing process. He was always available to answer my questions regardless of the size.

DEDICATION

To my family for all their unconditional support.

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CHAPTER I

INTRODUCTION

Motivations

With the proliferation of weapons technology to rogue nations like North Korea and Iran and the increasing prevalence of terrorism around the globe, there is growing concern that a nuclear device could be detonated in the United States or abroad as part of a terrorist incident. (1) As a result, there has been a renewed interest in nuclear forensics to be able to determine the origin and perpetrators of such a heinous act. (2) The forensics field because it is multi-faceted and includes a compilation of several different fields including chemistry, physics, and social science.

There is a basis for concern that a terrorist acquisition and use of a nuclear weapon may occur. The leader of Al Qaeda, Osama Bin Laden, has made many efforts to acquire nuclear weapons. The 9/11 Commission report sites that Osama Bin Laden's effort to develop nuclear weapons capabilities began sometime before 1994. (3) These efforts included attempts to buy uranium in Africa, Western Europe, and the former Soviet Union. (3) Bin Laden has also stated that the acquisition of weapons, including weapons of mass destruction, is a "religious duty" for all Muslims. (3) As a result, there is an obvious desire on the part of some sub-state organizations to acquire nuclear weapons, but perhaps more troubling is the potential for there to be suppliers of nuclear weapons related technology.

This thesis follows the style of *Nuclear Technology*.

The most egregious proliferation of weapons technology occurred in the A. Q. Khan nuclear black market. Khan was involved in both vertical (within a country) and horizontal proliferation (between countries). His horizontal proliferation included deals with Libya, North Korea, and Iran. (4) Even though the Khan network appears to be shut down, this nuclear know how has been sent around the world. The North Koreans are suspected of being similarly involved with spreading nuclear knowledge and abilities. The most recent mark against the Koreans is evidence and accusations by the United States that they were assisting Syria in building a graphite moderated reactor similar to the one at Yongbyon at a site known as Al-Kibar. (5) The site was destroyed by the Israeli Air Force in September 2007, so a conclusive determination of the activities at Al-Kibar may never be reached. However, the United States Intelligence community has implicated the North Koreans in playing a significant role in the Syrian effort. (6) The combination of the willingness to develop nuclear weapons side by side with the availability of knowledge is a troubling prospect.

Even though it would take a significant effort to develop a nuclear weapon covertly, it is a distinct possibility. Thus, preparations for the worst case scenario in which an unknown sub-state group detonates a nuclear device without warning must be prepared for. There are typically three different scenarios related to the weaponization of nuclear and radiological materials: Radiological Dispersal Devices (RDD), Nuclear Weapons, and Improvised Nuclear Devices (IND). This work is focused on the last scenario which is the most likely case for a homegrown nuclear device by a terrorist organization. Specifically, this work will focus on a plutonium-based IND.

Objectives: Statement of the Problem

This work focuses on a hypothetical situation where a terrorist organization has managed to detonate an IND. This means that the organization has managed to avoid detection while acquiring Pu, designing and manufacturing a viable device, and transporting their device to the place of attack. A plutonium-based IND would require approximately 8 kg (1 SQ as defined by the IAEA) of plutonium configured for an implosion style device.[7] These devices would be significantly less sophisticated than a typical state military device. The design would most likely be similar to that of the "Fat Man" bomb of the Manhattan Project.[8] Further, the yield of an IND should be well below 50 kilotons (kT). For an implosion device, the plutonium pit would be uniformly compressed with shaped explosive lenses in order to create a supercritical system.[9] The basic concept of an implosion weapon is illustrated in Figure 1.

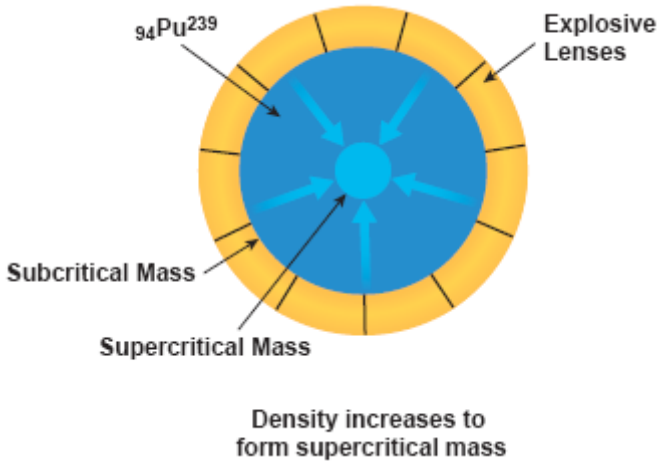

Figure 1: A Fat Man Style Implosion Concept[10]

Another basic variable which will have a significant impact on the scenario in question is related to the altitude of detonation. When the United States used its first nuclear weapons in the closing days of World War II, they were detonated at high altitude; however, it is more likely that a terrorist organization would use the device at ground level. The dispersal of fission products will depend greatly on the detonation altitude. In the high altitude scenario, the bomb fragments and fission products would be dispersed over a very large area. In the low altitude (and low yield) scenario or ground burst event, the fission products and device remnants would be scattered in the immediate vicinity of the explosion. This is the scenario considered in this work. Undoubtedly, this dispersion would not be uniform and a sampling procedure to best survey the area would need to be implemented in order to improve the accuracy of any method applied to the results of data taken from such samples.

The primary objective of this work was to produce a methodology that estimates pre-detonation isotopic ratios using post-detonation isotopic ratios measured with mass spectrometry of samples of the bomb debris. Sophisticated methods for this analysis have been developed by the U.S. national laboratories, but those methods are not discussed in the open literature. Accuracy of these methods is likely quite good, but computational speed may be limited. This work focused heavily on developing a method with an excellent computation speed even at the expense of accuracy. The intended use of the output of this model is to determine a reasonable initial estimation of the actual isotopics to be used in a more sophisticated and computationally intensive model. The ultimate goal of using this method as a pre-processer for other methods is to

significantly reduce the overall computational time for the attribution process and to provide results with sufficient accuracy to narrow the field of possible origins for the Pu used in the device. In the post-event attribution arena, being able to rapidly rule out specific sources is nearly as valuable as finding the exact solution.

Since much of the work in this field is not published in the open literature, a secondary objective of this work is to provide an open source analysis of the attribution problem. Countries who are considering transferring nuclear materials to groups with malicious intent may not fully comprehend the ability of U.S. scientists to trace the origin of materials back to their particular facilities. Thus, the open source nature of this work provides a deterrent to states considering transfer of nuclear materials to non-state actors. "[But,] nuclear attribution cannot succeed as a threat if the other side has no knowledge of it." (11)

Inverse and Forward Models

There are many potential complications and barriers to this research. Jay Davis, a founding director of the Defense Threat Reduction Agency (DTRA), is quoted saying:

> *I keep a standard mental list of the five hardest technical problems of which I am aware. Nuclear Forensics and Biological Forensics each make the list. There is no assurance that we can work backwards from the effects of these horrific events to uniquely determine a perpetrator.* (12)

This problem is difficult because it is an inverse problem in which we are attempting to estimate the original state of the system prior to detonation from measurements of the state of the system after detonation. An inverse problem is typically more complicated

than a forward problem. A forward model has specific inputs and characterizations of the materials in the original state and the physics describing the evolution of this state into the future is well known. This is not the case with the inverse problem. The amount of material needed to produce a viable nuclear weapon can range dramatically based on the sophistication of the design. "An important variable is the plutonium composition, a proliferant device is not likely to be designed or built with US stockpile grade plutonium."[13] Without the specifics of the design, there will be many unknowns that must be estimated including the amount of fissile material and packaging (e.g. transportation, disguise, etc.). Depending on the material and the energy spectra of the neutrons during the explosion, several physical properties of the system will change as a result of the explosion. The ^{239}Pu fission cross section is shown in Figure 2. These interaction cross sections describe the probability of a particular atom interacting with a neutron and can be guessed in an educated manner. "But if [the device] is improvised, that's less likely to work. It might not look like things you've seen before."[2]

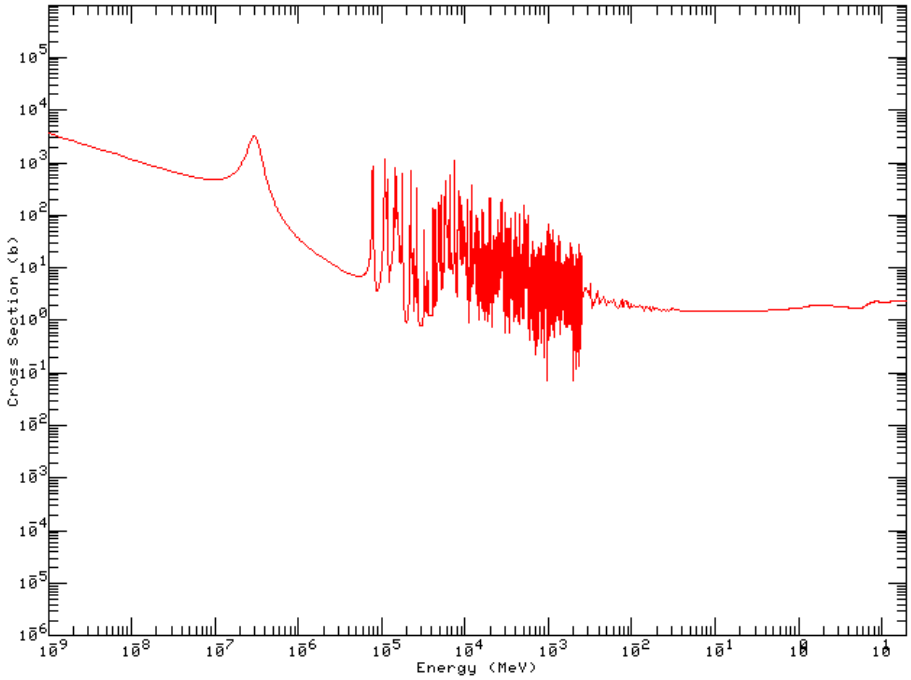

Figure 2: ^{239}Pu Fission Cross Section vs. Energy [14]

During a nuclear detonation, there will be hundreds of generations of neutrons interacting within the system with a population on the order of 10^{20} to 10^{22} neutrons. [10] The Pu atoms in the unexploded device will have the opportunity to undergo many competing reactions. During the explosion a single Pu atom potentially could interact several times creating a suite of isotopes which were not present in the unused device. There is no super computer which could model all of these interactions in a reasonable time frame. Thus approximations are made to speed up the process, and a resulting uncertainty is introduced.

The final complication is that the spatial distributions of reactions within the device will not occur uniformly. Different sections of the pit will experience a higher

neutron flux than others which in turn will cause different fission rates in various sections of the device. Upon the detonation of the device, much of ground zero will be vaporized only leaving miniscule particles to be collected and analyzed. The method of collecting a representative sample of the device is beyond the scope of this work; however, it should be noted that a methodology is only as good as its initial input data.

Hypothetical Scenario: Model Action Plan

For the hypothetical scenario outlined above, this work comes into play very late in the game of the nuclear forensics and attribution effort. The International Atomic Energy Agency (IAEA) has a model action plan to respond to these types of incidents. [15] The model action plan has set steps to ensure not only the safety of the people involved and control of nuclear material, but also the preservation of evidence. The action plan has guidelines from the initial response to the analytical follow-up. The following is a basic summary of what would occur onsite following any radiological emergency.

(1) Secure the Incident Site: The first and foremost objective is to ensure the safety and security of any people in the immediate vicinity of the incident including first responders.

(2) On-Site Analysis: This would include gamma-ray analysis, dose calculations, and a check for further hazards not initially noticed including secondary devices.

(3) Collection of Radioactive Evidence: After the site is deemed safe, a careful and comprehensive collection of evidence would occur according to accepted

radiological safety practices. Evidence could consist of sample swipes, liquid samples, soil samples, and other particles.

(4) Collection of Traditional Forensic Evidence: Some of the traditional evidence might have been destroyed during the collection of radioactive samples. Since the site will most likely still be contaminated, the people gathering evidence should practice good radiological safety to reduce dose and the further spread of contamination.

(5) Final Survey & Release of Scene: After all necessary forensic evidence has been collected, the site can be decontaminated if necessary and released to local authorities. [15]

After the onsite work has finished, there will be days to weeks of follow-up work done at the laboratory level. Ideally the analysis of the radioactive evidence would occur within the Network of Analytical Laboratories (NWAL). This could involve several different laboratories for quality assurance purposes. The working group suggests the formation of a team to manage the overall forensics effort. The analysis is highly dependent on the situation. They analysis would typically have two phases: basic characterization and full attribution. [15]

Basic characterization would be done fairly quickly upon receipt of the evidence. This type of analysis includes size, shape, mass, phase, molecular structure, grain size, gamma spectroscopy, and other non destructive analysis. The full attribution work that would follow includes computer modeling, mass spectrometry, x-ray fluorescence, and high resolution microscopy. Table 1 is a general timeline for the analysis following a

radiological incident. [15] The work described in this thesis would be part of the full attribution analysis step as outlined by Kristo et al. Ideally, this would be a good tool for obtaining preliminary results.

Table 1: Sequence for Forensic Tests after a Radiological Incident [15]

Techniques/methods	24 hours	One week	Two months
Radiological	Estimated total activity Dose rate (α, β, γ, n) Surface contamination		
Physical	Visual inspection Radiography Photography Weight Dimensions Optical microscopy Density	SEM/EDS XRD	TEM (EDX)
Traditional forensic	Fingerprints, fibres		
Isotope analysis	γ spectroscopy α spectroscopy	Mass spectrometry (SIMS, TIMS, ICP-MS)	Radiochemical separation
Elemental/chemical		ICP-MS XRF Assay (titration, IDMS)	GC-MS

Previous Work: Literature Review

The adaption of nuclear technology to solve problems related to nuclear forensics is not a new idea. The first example of nuclear forensics dates back to the first nuclear test by the Soviet Union in 1949. It was this analysis of airborne samples which conclusively proved that the Soviet Union had harnessed the power of the atom. [16]

However, based on recent concerns with nuclear terrorism, there has been a renewed interest in building modern nuclear forensics capabilities. The reemergence of the nuclear forensics program in the United States began in 1999.[11] As in the 1950's, the lion's share of this work has remained classified since it is unavoidably tied to sensitive nuclear weapons and national defense information. Resulting from this veil of secrecy, there is not an extensive library of unclassified science and policy literature available on this topic.

It is important to understand the differences between nuclear forensics and nuclear attribution. In the international community, the term nuclear forensics and nuclear attribution are often used interchangeably; however, for the purposes of this work, we will use the following definitions. Nuclear forensics spans across many different disciplines to utilize mass spectrometry, gamma spectrometry, metallurgy, electron microscopy, computer modeling, and much more. It includes the development, refinement, and application of technical means to examine nuclear material for uniquely identifying characteristics. Nuclear attribution is a specific use of forensics technology for many different problems including smuggling, unknown recovered sources, environmental sampling, contaminated areas, and verification of facility operations. Attribution also includes the political process in utilizing the scientific result on the world stage. Nuclear attribution borrows heavily from the forensics field, but it is the practice of applying nuclear forensic techniques in solving a specific problem. Techniques developed for forensics are utilized or adapted to assist in attribution efforts,

thus it is important to have a solid knowledge of forensic tools when working post-event attribution.

The United States scientific community is the leader in research related to forensics and attribution. Even though the technical data of such work is unavailable to the general public, there are reports related to exercises simulating the attribution process. One such simulation was undertaken in early 2004 where experts in fallout analysis gathered in Albuquerque, NM at Sandia National Laboratory. The scientists were divided into small teams and given US nuclear test data. The spokesman for the project commented that some teams succeeded in identifying the tests. [2]

Sandia National Laboratory is not the only Department of Energy organization working on the nuclear forensics and attribution effort. There are scientific research groups dedicated to the problem at both Los Alamos National Laboratory [17] (LANL) and Lawrence Livermore National Laboratory [18] [19] (LLNL). Ken Moody, one of the scientists working at LLNL, led a project to publish an academic textbook on nuclear forensics. The book focuses on understanding the processes in the nuclear field and how to detect them. Approximately three pages are focused on the post-event analysis of debris from either an RDD or IND event. The authors make it very clear that much of that work is classified and cannot be discussed in the book, but that the problem is being researched. [20]

One of the more recent advances, a high resolution gamma-ray detection system has been developed independently by both LANL and LLNL. The LANL microcalorimter has been able to provide gamma-ray spectra with up to a 47 eV

resolution at the full width at half max (FWHM) for low energy incident gammas. [21] The LLNL UltraSpec system has shown similar energy resolutions in the 50 to 200 eV range. The two differ in the method of cooling the system to tenths of a degree Kelvin. These two systems have been able to increase the resolution of the previous industry standard by approximately ten fold. As a result, these detectors distinguish between gamma-rays and x-rays in the same energy range. With this increase in detector resolution, more peaks related to special fissile material can be distinguished improving the accuracy of the calculation. Often in gamma-spectroscopy, the x-rays released by radiation interacting with metals surrounding the sample and the sample itself can obscure valuable peaks. These detectors could be used to more accurately, non-destructively measure the isotopics of fissile material and attributing natural uranium to a specific mine. [22]

The field of mass spectrometry has often been used in characterizing nuclear materials. This is by far the most accurate tool for determining isotopic ratios in a particular sample. It is typically the most expensive process in this series of analytical tools. Any variation of mass spectrometry will also destroy whatever sample it is analyzing. Thus, this technique is performed after some of the more standard non-destructive assay techniques such as gamma-ray spectroscopy. One example of mass spectrometry being used to solve the origin of unknown material is from Pacific Northwest National Laboratory (PNNL) at the Hanford site. Clean up crews came across a buried safe that contained a bottle of plutonium liquid. Jon Schwantes and his team at PNNL were able to track the material based on mass spectrometry analysis and

Department of Defense records to one of the very first plutonium separations at Oak Ridge National Laboratory (ORNL). They were even able to pin point the exact batch of fuel from which the plutonium was separated.

In addition to the work done on the national level, there have been two previous projects undertaken by students at Texas A&M University. The first of which was a project focused on the attribution of material in a Radiological Dispersal Device (RDD) event. This type of attack involves using conventional explosives to distribute radiological material. The work assumed spent nuclear fuel as the material used in the device. From various signatures, the work focuses on pinpointing the source of the spent fuel. The result of the work was the NEMASYS code which can identify several key characteristics of the material including burn-up, age, and enrichment within small uncertainty windows. [23]

The second effort conducted at Texas A&M was very similar to the current proposed research. However, highly enriched uranium, rather than plutonium was the fissile isotope used in the assumed IND. The Uranium analysis was undertaken by Adrienne LaFleur. She was able to use her methodology to trace HEU to a specific type of enrichment process. Further, she was able to determine a subset of mines from which the original ore was mined. Similarly, her project was undertaken with the speed of calculation as a foremost concern. [24]

Another group of leaders in the nuclear forensics field are located at the Institute for Transuranium Elements (ITU). They have been focused on the problem of illicit trafficking. This group uses a myriad of analytical tools including mass spectrometry

and electron microscopy to back track material through the black market. This group has been able to successfully determine the origin of plutonium to specific reactor types. [25] Their approach distinguishes between heavy water reactors, light water reactors, fast breeder reactors, and research reactors. But the similarities between the various classes of light water reactors does not allow for a determination between a PWR, BWR, or VVER. The same research group has also developed techniques for calculating the age of uranium [26] and age since separation of plutonium using isotope dilution mass spectroscopy (ID-MS) [27] [28]. This group assisted in the analytical work related to two seizures of illicit material in Europe and has published several case studies related to that work [29].

There are also several publications by the International Atomic Energy Agency (IAEA) and other organizations regarding the status of nuclear forensics capabilities. [30] One of the important publications by the IAEA provides International Target Values (ITV's). These are an evaluation of uncertainties used in various measurement techniques. The ITV's are considered to be the "state of the practice." These values are needed in producing quality assurance of measurement work being done around the world; however, it provides valuable insight into the technical capabilities of many of the laboratories around the world. Table 2 contains typical detection limits for measuring special nuclear material by various forensic methods. [31]

Table 2: Typical Detection Limits for Various Forensic Techniques [15]

Measurement goal	Technique	Type of information	Typical detection limit	Spatial resolution
Survey	HRGS	Isotopic	ng–µg	
Elemental and isotopic bulk analysis	Chemical assay	Elemental	Mg	
	Radiochemistry/RA Counting methods	Isotopic Elemental	fg–pg	
	TIMS	Isotopic Elemental	pg–ng	
	ICP-MS	Isotopic Elemental	pg–ng	
	GD-MS	Isotopic Elemental	0.1 ppb–10 ppm	
	XRF	Elemental	10 ppm	
	XRD	Molecular	~5 at%	
	GC-MS	Molecular	ppm	
	Infrared	Molecular	ppm	
Imaging	Visual inspection	Macroscopic	0.1 mm	
	Optical microscopy	Microscopic Structure		1 µm
	SEM			1.5 nm
	TEM			0.1 nm

These documents focus on the problem of solving the pre-explosion scenarios, but they still offer valuable insight into the possible analytical techniques and capabilities being used to tackle the difficult problem of post-event attribution. Many of the experts working on the projects listed above are also deeply involved with the efforts of the IAEA to provide nuclear forensic support in the case of an emergency. Thus, the available documents are similar in scope and content to much of the aforementioned projects. Even though the exact technical ability for attribution is not public knowledge,

by reading between the lines about the current work in mass spectrometry, high resolution gamma spectroscopy, and computer modeling, one discerns a significant effort toward developing nuclear forensics capabilities to be applied to nuclear attribution.

CHAPTER II

FORWARD MODEL THEORY

In this chapter, some background theory needed to understand the problem will be discussed. A more detailed description of the specific problem is followed by a primer on neutron induced reactions. This is followed by some basics of computer simulation of nuclear systems and the introduction of the forward model ORIGEN2.

IND Basics

There are two basic designs for an improvised nuclear device: gun-type assembly or implosion type assembly. Gun-type weapons must be made using Highly Enriched Uranium (HEU), and implosion type weapons could be made using either Pu or HEU. This work is solely concerned with plutonium based devices. Thus, only implosion weapons will be considered. When using Pu as the fissile material for the device, a gun-type assembly would not yield a viable device due to the high spontaneous fission rate of ^{240}Pu – the main contaminant in any Pu sample. Due to the increased mass required to build a gun-type device coupled with the spontaneous fission from ^{240}Pu, a gun-type weapon utilizing Pu has a very high chance of pre-detonation or a fizzle should the device make it to its intended target. [32]

Some of the basic components expected to be seen in a viable implosion IND are the fissile pit, fast and slow explosives, electronic controls, and packaging. Depending on the sophistication of the design, a tamper, reflector, and initiator could also be present. For the purposes of this work, the sole concern is the fissile plutonium pit; however, for the overall attribution effort, many clues related to finding the culprits

could be found by analyzing the other portions of the bomb if they survive the blast. Implosion weapons can also experience fizzle detonations as well.

When a device does not achieve its designed yield, it is called a fizzle. There are two common causes for a fizzle: non uniform compression of the fissile pit or slow assembly of the supercritical mass. [33]

Computer Modeling

Due to a lack of available sample data, this work will need to utilize a forward model to produce input data equivalent to what would be measured by mass spectrometry analysis of samples. As previously discussed, a weapon and a nuclear reactor generally obey the same basic laws of nuclear physics. Fortunately, there has been a significant amount of work on the development of reactor simulation. This work will use a previously existing nuclear reactor physics code for the forward model. There is a multitude of codes that can be used to track production, depletion, and decay of plutonium and actinide isotopes in a nuclear reactor core. These codes have been developed by the national laboratories, academic institutions, and individual nuclear reactor vendors.

The two most common types of codes are Monte Carlo and deterministic. The Monte Carlo method tracks individual particles from birth to death while using random numbers and probabilities to predict nuclear interactions. Since this is a statistical process, the more particles tracked, the better the results. But as a consequence, it is common for millions of particles to be run in one simulation which takes a significant amount of computational time. The most common and widely used code of this type is

the Monte Carlo Neutral Particle (MCNP) code developed by Los Alamos National Laboratory. [34] The MCNP code has several incarnations with different abilities and features. Typically the MCNP suite of codes is considered to be extremely accurate and is the main benchmarking resource in the nuclear research and development industry.

Deterministic codes solve specific equations as opposed to following specific particles from birth to death. In nuclear engineering, deterministic codes typically solve either the transport equation or the diffusion equation. These codes vary in complexity and have incarnations that address problems spanning from three dimensional multi-group solutions to one dimensional single-group solutions. One example of a deterministic code is TransLAT. TransLAT has the ability to solve one to three dimensional problems as well as multi-energy-group problems. [35]

Another deterministic option would be to run a lattice physics code. HELIOS is one version of this type of code. This code solves the neutron transport equation using the method of angularly dependent, current-coupled collision probabilities. HELIOS solves multi-group problems with up to 190 neutron energy groups. [36] HELIOS has been benchmarked to different power reactor types and performed very well compared to experimental measurements of spent fuel. [37] The problem with using a lattice physics code is that it depends on having a large number of repeated cells. Typically when modeling a reactor in a lattice physics code, one fuel pin will be modeled and the results of that simulation will be extrapolated to the full reactor size. This type of modeling is not appropriate for the IND problem since the specifics about the IND design are most likely unknown.

Each specific code has different methods of solving the governing equations which introduces different embedded assumptions. An example of these embedded assumptions is that in reality the transport equation is intrinsically coupled with the change rate equations for a given system. As time passes, the number of atoms of a specific isotope will be changing as well as the incident neutron flux. Thus the reaction rates for each isotope will be changing with time. This time dependence exponentially increases the complexity of the system of equations to be solved. A standard way of simplifying this time dependence is to decouple the transport and change rate equations by using a constant flux.

Another type of code that would be very appropriate for this work is a nuclear weapon simulation code. However, these codes are not available for unclassified public use.

The Forward Model: ORIGEN2

The forward model selected for this work was the Oak Ridge Isotope Generation and Depletion Code Version 2.2 or ORIGEN2. ORIGEN2 was developed in the 1980's and 1990's to calculate the buildup, decay, and processing of radioactive materials. "ORIGEN2 uses a matrix exponential method to solve a large system of coupled, linear, first-order ordinary differential equations with constant coefficients." [38] This is a very computationally efficient program and runs are typically less than one minute on a modern personal computer.

The ORIGEN2 code is an open source code that can be accessed by the general public. Typically ORIGEN2 is used for modeling pressurized and boiling water reactors

as well as reprocessing facilities, but the code has the ability to model an extensive number of reactor types and situations. This particular code has several advantages. One of the reasons for choosing ORIGEN2 is related to its library for the Fast Flux Test Facility (FFTF (38)) for plutonium and uranium. This library provides the one group fast neutron interaction cross sections and fission yields for both plutonium and uranium. These cross sections are read into ORIGEN2 from a preexisting library. Other codes create collapsed cross sections from data based on the input deck. During execution, the program tracks the probabilities of changes in the material with a transition matrix, this matrix is built based on the input parameters and consists mostly of zeros to form the matrix for the exponential matrix method. ORIGEN2 uses a composite solution algorithm composed of three different techniques. First the program uses asymptotic solutions to reduce the transition matrix to a more manageable size. This technique effectively removes the shortest lived products from the model. Then the reduced transition matrix is further reduced by the exponential matrix method. The final phase is to use a Gauss-Seidel successive algorithm in conjunction with another set of asymptotic solutions. More specifics about the computational workings of ORIGEN2 can be found in the ORIGEN2 Manual listed in the references section. [38] [39]

ORIGEN2 treats the system as zero-dimensional, point-wise, and homogeneous. This type of model averages the incident flux and interactions over the entire system. This is appropriate for this work since the exact design and dimensions of the IND are not known, and it is possible to add uncertainty by incorrectly making assumptions regarding the design. A more sophisticated forward model would require a significantly

larger amount of detail regarding the geometry and material compositions comparatively. ORIGEN2 also can easily calculate the radioactive decay of the fission products in a single step following irradiation. As a result, the output of ORIGEN2 can be fed directly into the inverse model. Even though ORIGEN2 is not a weapons simulation code, it has the capability to provide data to study the feasibility of this methodology. [24]

As with any forward model, ORIGEN2 has a few potential issues. Since this code was not developed to model nuclear weapons, the model could have issues with the extremely short time period of a nuclear detonation. One further issue is that the ORIGEN2 code is fairly old in the world of computer simulation. There has not been any updating of the code since 2002. [38] As a result, ORIGEN2 might not be using the most up to date modeling techniques or nuclear data. Despite those potential issues, ORIGEN2 is an appropriate forward model to use with this inverse model because of its versatility, computational efficiency, performance in benchmarking and wide spread use.

CHAPTER III

INVERSE MODEL DERIVATION

The approach of this methodology is to start from first principles of nuclear interactions to derive a system of equations to estimate the pre-detonation isotopics of the fissile material originally used in the IND based on the isotopics of key nuclides after the detonation. The input data (post-detonation isotopics) for this system would come from mass spectrometry analysis of samples taken around the blast site. Mass spectrometry analysis typically results in ratios of atoms of a particular isotope to another reference isotope. Some discussion of how the inverse model was derived is given in this chapter along with the resulting equations. A full step-by-step derivation of the system of equations can be found in APPENDIX B.

Simplifying Assumptions

The actual nuclear physics occurring in this system is particularly complicated. An even further complication is that the device being modeled for this inverse methodology has a significant number of unknowns. It would take an enormous amount of computing power and time to attempt to model the processes without making some simplifying assumptions. One of the key objectives of this work is the development of a computationally efficient model. The following assumptions are embedded in this inverse model.

During the short lifetime of the nuclear detonation, the neutron flux within the device will vary by region and time. In order to remove the time dependent nature of the neutron flux, a time and spatially averaged neutron flux was used. This particular

assumption is very common in nuclear engineering forward burn codes. Each isotope could experience several different interactions with the incident neutrons. But each isotope will have a dominant production and depletion interaction. These neutron induced interactions will physically alter the material on the atomic structure. Many of the competing reactions will not change the material, so they can be ignored in this work.

Further, any particular atom could undergo several interactions, but the total time of the nuclear detonation will be on the order of 10^{-6} seconds. This is much shorter than in a nuclear reactor, so in order to remove several small terms at the end of the change rate equation this work will assume that each atom will undergo only one interaction during the detonation. Since the amount of material depletion and production is relatively small, this is not an unrealistic assumption. The amount of each isotope will change during the event, but the changes are less than one order of magnitude.

Another assumption comes out of the time scale of the nuclear event. Since the fission products that will be used in this methodology have long half-lives compared to the event time frame, the infinitesimally small number of decays occurring during the detonation will be neglected. This assumption introduces little error due to the fission product selection that will be discussed later, but the decay occurring during the time between detonation and sample analysis will be addressed. Further, the isotopes of plutonium being considered in this system are radioactive; however, their half-lives are extremely long. Radioactive decay of plutonium atoms in general during the entire process, detonation to sample analysis, will be ignored completely.

The final computational assumption involves the neutron interaction cross sections. The cross sections for each isotope vary based on the incident neutron energy. Running an energy dependent neutron code would significantly increase the overall computation time. Thus, the fission averaged cross section data from the ORIGEN2 library will be used. Since all of the neutrons appearing after the first generation can be assumed to be produced by fission, this is not an uncommon assumption. Even though the number of assumptions embedded in the derivation of this methodology seems high, they are common nuclear engineering approximations which are well understood in the reactor modeling field.

Atomic Level Production and Depletion

The basic nuclear physics of a nuclear explosion is not much different than that of a nuclear power reactor. On the atomic level, neutrons are interacting with the individual atoms in the material. Each material will interact differently based on various quantum mechanical properties of the atomic structure. These different interactions are quantified into nuclear cross sections, which are essentially probabilities that a certain reaction will occur for a specific atom for a neutron track length and number of incident particles. These cross sections depend on various parameters including the energy of the incident particle and temperature of the material. For this work, fast neutrons are the main concern since no moderator is present in the weapon.

In the relatively short lifetime of a detonating IND, isotopes will be depleted and will build up within the system. In this particular system, the primary isotope being depleted will be ^{239}Pu. This will produce fission products and higher actinides. Since

this work focuses on the pre-detonation characterization of the material, neutron induced production and depletion reactions are paramount.

Scattering and Absorption

When a neutron comes into contact with an atom, there are two possible outcomes: scattering or absorption. Scattering reactions do not result in production or depletion of nuclei. The other possibility is that the neutron will be absorbed by the nucleus. Absorption reactions result in depletion or production of nuclei. Absorptions reactions include fission, radiative capture, neutron production, and charged particle production. The probabilities for some of these reactions are very small compared to competing reactions.

Properties of Plutonium

Since this work is only concerned with IND's composed of plutonium, the isotopes of concern will be ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, and ^{241}Am. Each of these isotopes is likely to be present in any plutonium pit. Thus, each carries information related to the origin of the material. All of these isotopes will be subject to a flux of neutrons causing build up and depletion. The work described in the literature review centered on tracing material that had not been detonated – pre-event attribution. But the materials and isotopic ratios will change as the neutrons interact with the IND atoms during the event. Using these interaction probabilities, the post-event material isotopics can be used to produce pre-event isotopics which can take advantage of previous efforts to attribute the origin or source of the material.

Each isotope will interact similarly; however, each will have different interaction tendencies. The total cross sections of the plutonium isotopes are on the same range, but each has unique probabilities for each interaction. For example, ^{239}Pu is less likely to scatter a neutron than ^{240}Pu, and ^{241}Pu is five times more likely to have a neutron producing reaction than ^{239}Pu. The neutron interaction cross sections for these isotopes are given in Table 3.

Table 3: Cross Section Properties of Plutonium [14]

Nuclide	Total	Fission (n,f)	Radiative Cap. (n,γ)	Scattering (n,n)	Neutron Producing (n,2n)
^{238}Pu	8.829	1.994	0.0993	6.7306	0.003669
^{239}Pu	7.712	1.80	0.05294	5.854	0.004045
^{240}Pu	7.723	1.357	0.09328	6.269	0.003547
^{241}Pu	7.84	1.648	0.1182	6.052	0.02137
^{242}Pu	7.934	0.08787	1.127	6.71	0.006667
^{241}Am	7.784	1.378	0.2296	6.173	0.0006204

All cross sections are the fission spectrum average
All cross sections in barns

Dominant Production and Depletion Mechanisms

For the inverse model, some of the interactions can be ignored. There are many different types of reactions occurring simultaneously during a nuclear event. Often, the most likely reaction, may not add detail to the methodology. For example, ^{238}Pu will undergo some fissions in this system; however, due to the small proportion of this particular isotope, these fissions will not significantly contribute to the fission product signature. The production and depletion mechanisms in this system are radiative capture to create actinides higher than ^{239}Pu, the fission of ^{239}Pu, and the (n,2n) neutron

producing reaction in ^{239}Pu. The dominant production and depletion interactions are seen in Figure 3.

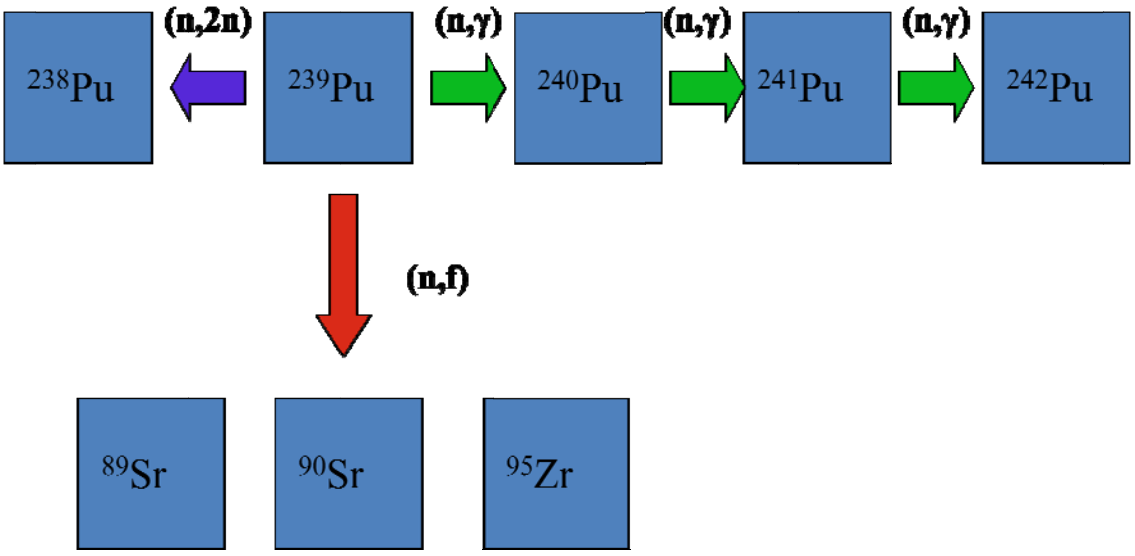

Figure 3: Dominant Production and Depletion Mechanisms

Burn-up and Yield

The main physical quantity related to the production of all isotopes in this system is the scalar flux of neutrons. Both fission products and higher actinide production are directly related to the total flux of neutrons within the device. The total flux is related to the overall yield or burn up of the material. The yield per unit volume (Ψ) is equal to the integral of the macroscopic fission cross-section of ^{239}Pu multiplied by the energy produced per fission and the neutron flux with respect to time:

$$E_r \int_0^T \Sigma_f^{Pu\text{-}239}(t)\phi(t)dt = \Psi\rho^{Pu\text{-}239} \quad (1)$$

where (E_r) is the energy recoverable from fission, and ($\rho^{Pu\text{-}239}$) is the density of ^{239}Pu, and where $\Sigma_f^{Pu\text{-}239}$ is the macroscopic fission cross-section of ^{239}Pu.

This approximation of the yield will tie all of the equations for different isotopic ratios together. In order to simplify the equations, a burn-up factor (Ψ_X) will be used. This factor, which is the yield multiplied by a physical constant, is given by,

$$\Psi_X = \Psi \frac{N_{Pu\text{-}239}(0)}{N_{Pu\text{-}239}(T)} \frac{N_{Pu}(0)}{N_{Pu\text{-}239}(0)} \quad (2)$$

where $N_X(t)$ denotes atom density of isotope X at time t.

For this work, it is important to note that the quantity being calculated is essentially the total number of fissions. An approximate yield is found by assuming the device is made from one significant quantity as defined by the IAEA. The significant quantity for plutonium is 8 kg. So the yield is essentially energy released for a given amount of mass. Burn-up is a similar quantity. Burn-up is used in quantifying the amount of energy released for a specified amount of material in reactors. These two normalized groups are intrinsically related. In this work, they are considered to be the same value.

Fission Product Selection

One of the main products that will build up in the bomb's fissile core will be fission products. These are the two heavy masses produced by splitting the ^{239}Pu atom. In any workable nuclear device design, there should be effectively no fission products present in the material before detonation. Thus, all of the fission products will have been

a direct product of the neutron flux seen during detonation. From the fission products, a value for the energy released can be approximated.

There are many possible fission products to be considered for use in this methodology. The three fission products selected for this work are: ^{89}Sr, ^{90}Sr, and ^{95}Zr. These three fission products have been selected based on their nuclear properties. An important reason for their selection is that they have similar atomic weights which will be advantages for a mass spectrometry analysis. Also, these particular mass chains have relatively high cumulative yields than some other fission product mass chains. The mass-yield chart for the fission of plutonium is shown in Figure 4. The yield for these isotopes is different for the fission of ^{235}U, but these isotopes are prominent in that process as well. Further, each of these fission products has a relatively long half life. Most fission products are very short lived and decay rapidly toward more stable isotopes. Each of these isotopes is near the line of stability on the chart of the nuclides. The final advantage these particular isotopes have is a small neutron absorption cross section in the fission averaged spectrum. This means that the fission products produced in the event will not significantly "burn" out of the sample during the event. [14] [40] Since the probability of one of these fission products undergoing a reaction is small, this work assumes that every fission product produced will remain after the detonation. The physical constants for these fission products can be seen in Table 4.

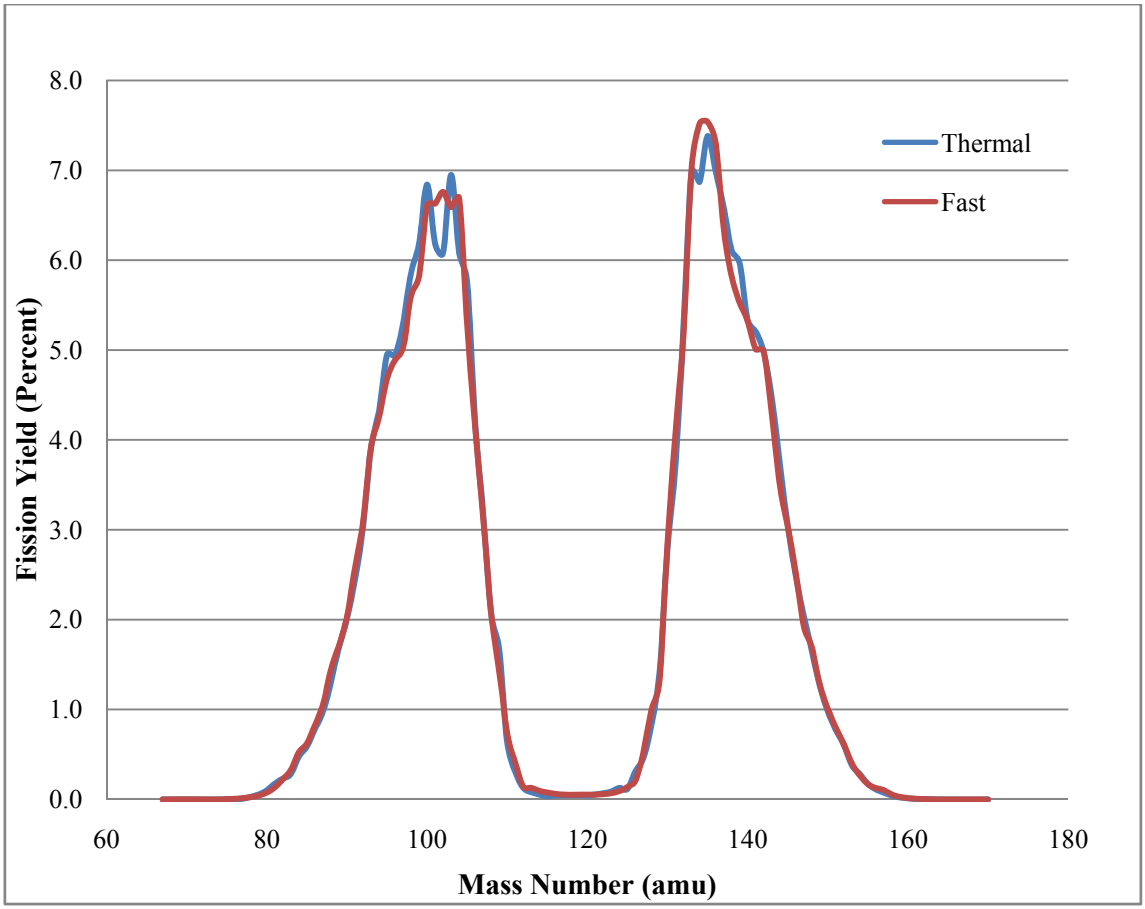

Figure 4: Fission Yield vs. Mass for ^{239}Pu [41]

Table 4: Fission Product Properties [14]

Fission Product	Half Life	Cum. Yield (percent)	Total Cross Section (barns)	Absorption (barns)	Scattering (barns)
^{89}Sr	50.52 days	1.81	5.662	0.00245	5.647
^{90}Sr	28.78 years	2.13	5.661	0.00421	5.652
^{95}Zr	64.02 days	4.77	5.661	0.02721	5.622

Time of Explosion (Decay Time Correction)

Undoubtedly, there will be a time gap between the detonation of the device and the sample analysis. The fission product decay occurring during this time gap must be

accounted for. The exact time since detonation can be found by using measured samples of $^{89}Y/^{239}Pu$ and $^{89}Sr/^{239}Pu$. The fission yield of ^{89}Y is very small, and it is a reasonable assumption to set the initial value of ^{89}Y after detonation to zero. Also, ^{89}Y is stable [42], and will not decay further. Thus any ^{89}Y will have grown into the samples by radioactive decay of ^{89}Sr. This decay time (T_{decay}) is given by

$$T_{decay} = \frac{1}{\lambda_{Sr\text{-}89}} \ln\left[\frac{N_{Y\text{-}89}(T)}{N_{Sr\text{-}89}(T)} + 1\right] \quad (3)$$

where $\lambda_{Sr\text{-}89}$ is the decay constant for ^{89}Sr.

The exact time of detonation will be recorded by other means allowing an opportunity to compare the calculated detonation time with the observed detonation time. This gives a check to see if the samples taken have produced credible data.

Ψ_X Calculation with Fission Products

After each measured fission product isotopic ratio has been corrected for the decay experienced between the event and measurement, the three selected fission products can be used to estimate the overall energy released from fission. Since the absorption cross-section for each fission product is small, the depletion of fission products due to neutron absorption has been ignored. Thus each fission product has one simple equation to relate the measurements to the burn-up of the material. An average of the three results will yield the best approximation for the burn-up factor. The equations are:

$$\Psi_X = \frac{N_{Sr\text{-}89}(T)}{N_{Pu\text{-}239}(T)} \frac{E_r N_a}{Y_{Sr\text{-}89} m_{Pu}} \quad (4)$$

$$\Psi_X = \frac{N_{Sr\text{-}90}(T)}{N_{Pu\text{-}239}(T)} \frac{E_r N_a}{Y_{Sr\text{-}90} m_{Pu}} \quad (5)$$

$$\Psi_X = \frac{N_{Zr\text{-}95}(T)}{N_{Pu\text{-}239}(T)} \frac{E_r N_a}{Y_{Zr\text{-}95} m_{Pu}} \quad (6)$$

where Y_x is the cumulative fission yield of isotope X, N_a is Avogadro's number, and m_{Pu} is the original atomic mass of Pu. The three equations are basically the same with the corresponding cumulative fission yields for each isotope.

Plutonium Ratio Calculations

Ultimately to trace IND material to its origin, information must be gathered from all the materials in the device including any contaminants. So, this work is concerned with determining the isotopic ratios of all the major plutonium isotopes that could have originally been in the system. These isotopes are the range between ^{238}Pu and ^{242}Pu, including ^{241}Am. The equations for these isotopes are all solved successively using the burn-up factor found from the fission products and the result from the previous equation. Each isotope is assumed to have only one dominant reaction in order to simplify the equations. These reactions are discussed in Chapter II. To further simplify the equation, each atom is assumed to only undergo one interaction. This assumption means that only the atoms present in the original material can be transmuted into other isotopes.

The ratio of ^{239}Pu atoms before detonation to ^{239}Pu atoms after detonation is given by

$$\frac{N_{Pu\text{-}239}(0)}{N_{Pu\text{-}239}(T)} = 1 + \frac{\sigma_a^{Pu\text{-}239}}{\sigma_f^{Pu\text{-}239}} \frac{m_{Pu}}{N_a} \frac{1}{E_r} \Psi_X \quad (7)$$

where $\sigma_a^{Pu\text{-}239}$ is the microscopic absorption cross-section of ^{239}Pu, $\sigma_f^{Pu\text{-}239}$ is the microscopic fission cross-section of ^{239}Pu.

Each other isotope of interest has an equation to determine its pre-detonation isotopic ratio with the post-detonation atom density of ^{239}Pu. The equation for ^{238}Pu is slightly different from the other plutonium equations. This is due to the production mechanism. ^{238}Pu is created from ^{239}Pu by an (n,2n) neutron producing reaction. The main difference in the equation is that the cross sections have been changed to reflect this difference. These ratios are given by:

$$\frac{N_{Pu\text{-}240}(0)}{N_{Pu\text{-}239}(T)} = \frac{N_{Pu\text{-}240}(T)}{N_{Pu\text{-}239}(T)} - \frac{\sigma_\gamma^{Pu\text{-}239}}{\sigma_f^{Pu\text{-}239}} \frac{m_{Pu}}{N_a} \frac{1}{E_r} \Psi_X \tag{8}$$

$$\frac{N_{Pu\text{-}241}(0)}{N_{Pu\text{-}239}(T)} = \frac{N_{Pu\text{-}241}(T)}{N_{Pu\text{-}239}(T)} - \frac{N_{Pu\text{-}240}(0)}{N_{Pu\text{-}239}(T)} \frac{\sigma_\gamma^{Pu\text{-}240}}{\sigma_f^{Pu\text{-}239}} \frac{m_{Pu}}{N_a} \frac{1}{E_r} \Psi_X \tag{9}$$

$$\frac{N_{Pu\text{-}242}(0)}{N_{Pu\text{-}239}(T)} = \frac{N_{Pu\text{-}242}(T)}{N_{Pu\text{-}239}(T)} - \frac{N_{Pu\text{-}241}(0)}{N_{Pu\text{-}239}(T)} \frac{\sigma_\gamma^{Pu\text{-}241}}{\sigma_f^{Pu\text{-}239}} \frac{m_{Pu}}{N_a} \frac{1}{E_r} \Psi_X \tag{10}$$

$$\frac{N_{Pu\text{-}238}(0)}{N_{Pu\text{-}239}(T)} = \frac{N_{Pu\text{-}238}(T)}{N_{Pu\text{-}239}(T)} - \frac{\sigma_{n,2n}^{Pu\text{-}239}}{\sigma_f^{Pu\text{-}239}} \frac{1}{E_r} \frac{m_{Pu}}{N_a} \Psi_X \tag{11}$$

where $\sigma_\gamma^{Pu\text{-}239}$ is the microscopic (n,γ) absorption cross-section for ^{239}Pu, $\sigma_\gamma^{Pu\text{-}241}$ is the microscopic (n,γ) absorption cross-section for ^{241}Pu.

Americium-241 is a very important isotope in nuclear forensics and attribution. When plutonium is freshly separated, there is no ^{241}Am present. But in plutonium samples that have aged, ^{241}Am builds up as ^{241}Pu decays by beta particle emission. Thus the relative quantity of ^{241}Am present is related to time since the last reprocessing of the plutonium sample. Using the time since the last reprocessing is a way to eliminate many

possible sources for the material. So this work methodology also includes a calculation of the ^{241}Am ratio. This particular isotope is not produced during the nuclear chain reaction, but it will be depleted slightly by an incident neutron flux. By using the same assumptions used for computing the ratios of plutonium isotopes, the amount of ^{241}Am present at the instant before the event can be calculated by

$$\frac{N_{\text{Am-241}}(0)}{N_{\text{Pu-239}}(T)} = \frac{N_{\text{Am-241}}(T)}{N_{\text{Pu-239}}(T)} \exp\left[\frac{\sigma_{\text{absorb}}^{\text{Am-241}}}{\sigma_{\text{f}}^{\text{Pu-239}}} \frac{m_{\text{Pu}}}{N_a} \frac{1}{E_r} \Psi_X\right] \tag{12}$$

where $\sigma_{absorb}^{Am\text{-}241}$ is the microscopic (n,γ) absorption cross-section for ^{241}Am.

After determining the ratios of all the Plutonium isotopes with respect to the post-event ^{239}Pu number density, the total number of fissions can be found. This is done by summing the plutonium isotope ratios and dividing this into the burn-up factor.

$$\Psi = \Psi_X \frac{N_{\text{Pu-239}}(T)}{N_{\text{Pu}}(0)} = \Psi_X \sum_{\text{isotopes}} 1 \Big/ \frac{N_{\text{isotope}}(0)}{N_{\text{Pu-239}}(T)} \tag{13}$$

The initial ^{240}Pu/^{239}Pu ratio is found using:

$$\frac{N_{\text{Pu-240}}(0)}{N_{\text{Pu-239}}(0)} = \frac{N_{\text{Pu-240}}(0)}{N_{\text{Pu-239}}(T)} \Big/ \frac{N_{\text{Pu-239}}(0)}{N_{\text{Pu-239}}(T)} \tag{14}$$

Similarly for all isotopes of interest the pre-detonation ratio can be found with:

$$\frac{N_{\text{Isotope}}(0)}{N_{\text{Pu-239}}(0)} = \frac{N_{\text{Isotope}}(0)}{N_{\text{Pu-239}}(T)} \Big/ \frac{N_{\text{Pu-239}}(0)}{N_{\text{Pu-239}}(T)} \tag{15}$$

These are the full isotopic ratios of the pre-detonation Plutonium.

Use of Methodology (Flow Chart)

The system of equations derived above has many interrelated parts. A flow chart of the implementation of the methodology can be found in Figure 5. The sample data

created by ORIGEN2 is first used to find the decay time and each of the fission products are then adjusted to reflect the calculated amount of decay time. This is done using Equation 3. The three fission product ratios are used to determine the burn-up factor, Ψ_X. The results from Equations 4, 5, and 6 are averaged to get Ψ_X. From here, the ratio between ^{239}Pu before and after detonation is found. This is done using Equation 7. This ratio in combination with the burn-up factor is used to find all the ratios for the remaining plutonium isotopes using equations 8 – 12. The results from the entire suite of isotopes are recombined to find the yield of the device with Equation 13. Concurrently, each ratio is also corrected to the pre-detonation time frame using Equations 14 and 15.

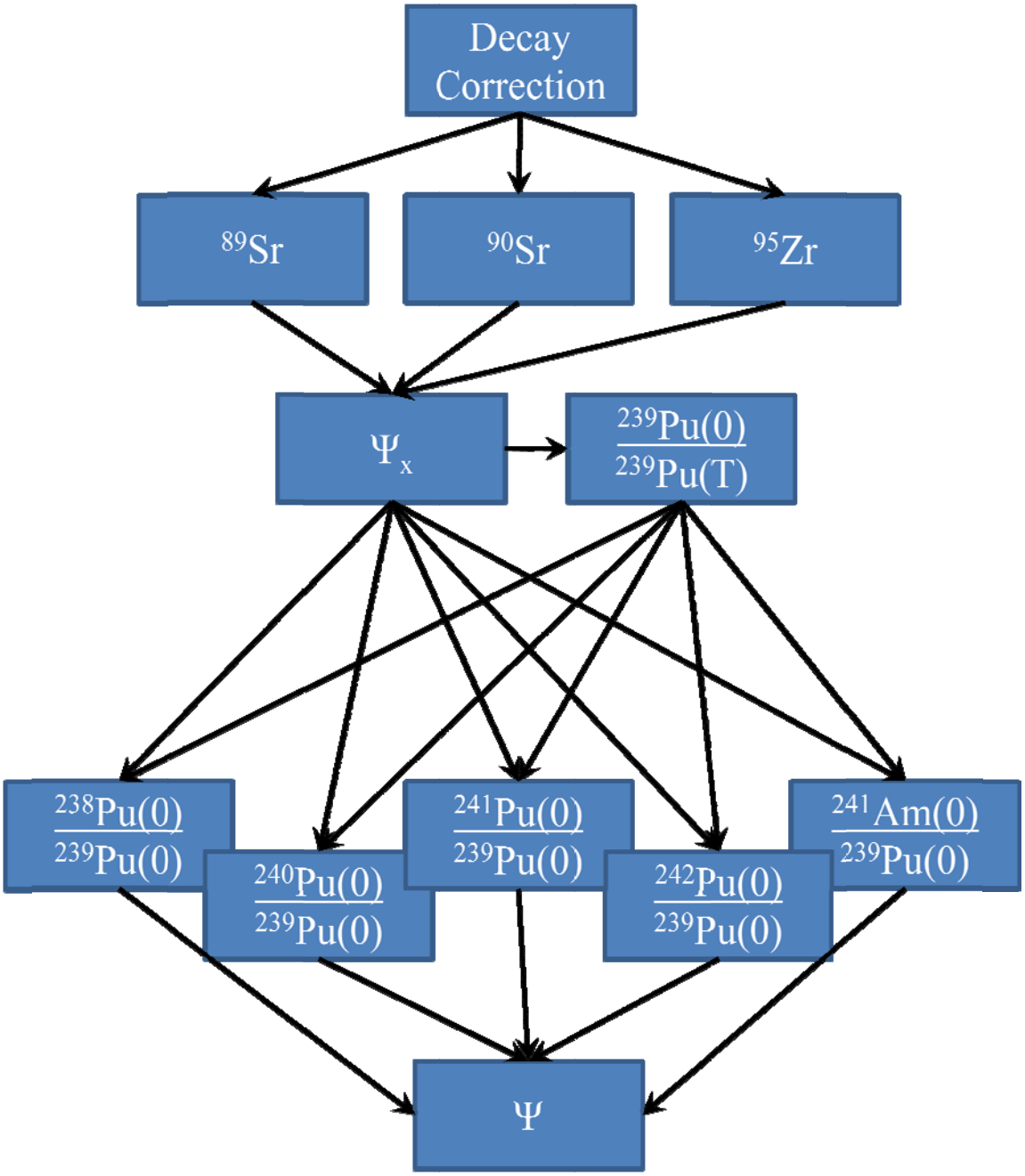

Figure 5: Inverse Methodology Flow Chart

CHAPTER IV

RESULTS AND DISCUSSION

Presented here are the results from testing the feasibility of the methodology. The discussion begins with the input sample selection, followed by the testing with the forward model, the inverse model execution, and a discussion of the sensitivity of the methodology.

Input Samples

As previously discussed, there is a broad spectrum of plutonium isotopics that could be used to create a viable improvised nuclear device. In order to best test the range of possibilities, several different scenarios were used. The testing was broken down into two main cases: simplified and realistic. In the simplified, there are only two nuclides present in the bomb material: ^{239}Pu (95 weight percent) and ^{240}Pu (5 weight percent). In the realistic cases, the other isotopes of plutonium and ^{241}Am included. Both weapons grade plutonium and reactor grade [43] plutonium are considered in the realistic cases. The weight percent of each isotope used in the cases are listed in Table 5.

The simplified case is designed to test the response of the methodology over various yields of devices. It is impossible to say what the exact yield a terrorist organization would attempt to achieve; however, the most likely scenario is that a terrorist device would be well under 50 kilotons (kT). The realistic cases are designed to test the performance of the methodology with an entire suite of plutonium isotopes present. With the increase plutonium "contaminants," there will be more of the competing reactions related to the higher plutonium isotopes. Since there is debate on

whether or not a functional bomb can be made out of reactor grade plutonium, a reactor grade sample has been included. The purpose of this is two-fold. This sample has a significant amount of contaminants and it is at the lowest end of ^{239}Pu content which covers the full range of possibilities.

Table 5: Plutonium Samples Used with ORIGEN2

Nuclide	Ideal World weight percent	Weapons Grade weight percent	Reactor Grade weight percent
^{234}U	0.0	0.00046	0.0
^{237}Np	0.0	0.00088	0.0
^{238}Pu	0.0	0.00498	2.87
^{239}Pu	95.0	94.98	54.34
^{240}Pu	5.0	4.8	25.72
^{241}Pu	0.0	0.123	7.05
^{242}Pu	0.0	0.0052	6.72
^{241}Am	0.0	0.089	3.3

Forward Model Results

Before jumping to the testing the inverse model, several tests were run on the forward model. Since ORIGEN2 uses a homogeneous cross sections and fission yields, users of ORIGEN2 often use multiple irradiation periods in the input deck to more realistically account for the changes of material and neutron flux over time. (39) However, this work has an extraordinarily short irradiation time as compared to a reactor and a multiple step irradiation is not practical. To test the response of ORIGEN2 over short time periods a series of runs was completed. The input material was the Weapons Grade plutonium sample described above. This simulation was for a ten kiloton blast. The irradiation time was varied from a half second and very high neutron flux to six

thousand seconds and proportionally smaller neutron flux. The result was that the code calculated the exact same solution at the low time interval as the high time intervals. The isotopic ratios for the selected fission products and ^{240}Pu with respect to ^{239}Pu output by ORIGEN2 during these tests are shown in Figure 6. Due to the consistency of the results, the ORIGEN2 output will not vary as a result of the short irradiation time step.

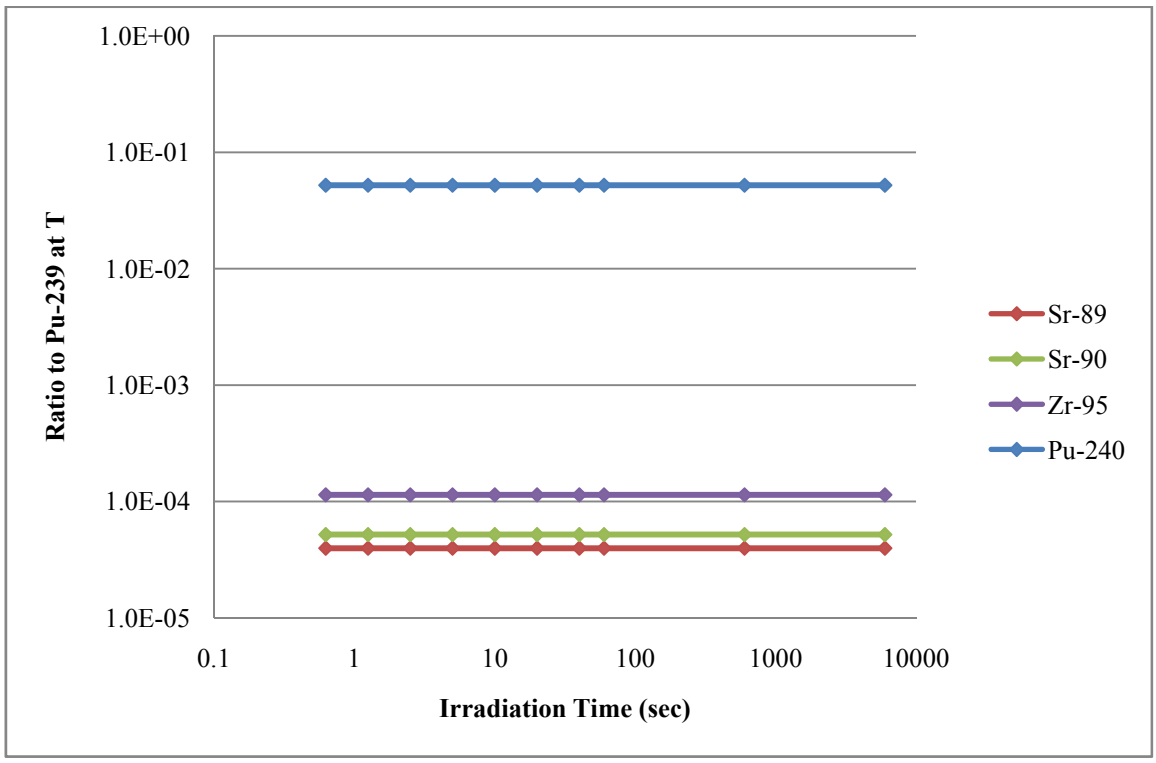

Figure 6: Irradiation Time Interval Test Results for ORIGEN2

Also, the ORIGEN2 code has two different versions. One is designed for thermal reactors and the other is designed for fast reactors. During the time interval test, the two versions of ORIGEN2 were compared. For the same input materials, irradiation times, and decay times, the two codes performed identically. Since the two codes are

solving the same physics equations while using identical cross sections and fission yields, the identical performance of the two codes is predictable. Despite the congruence of the two programs, the ORIGEN2 version designed for fast reactors will be used for the remainder of this work.

Inverse Model Results

Our inverse model equations were programmed into Microsoft Excel. From the forward model, the grams of each isotope are uploaded into the program where isotopic ratios are calculated. Using these, the system of equations runs within Excel exceptionally quickly. A big advantage for this implementation is that each step can explicitly be seen in order to trace any inconsistencies.

Simplified Case

The simplified scenario was tested over a broad range of possible weapons yields starting from 0.1 kT to 100 kT. The range of yields was incremented by five kT intervals. Since this scenario contains only two plutonium isotopes, the equations for the other plutonium isotopes are not used. As a result, the simplified case only has two solutions. These solutions are the yield and the ^{240}Pu to ^{239}Pu ratio. The solution given by the methodology was compared to the expected results as was given by the forward model. The two numbers were compared using the percent difference formula,

$$\text{Percent Difference} = \frac{|N_{actual} - N_{calculated}|}{N_{actual}} * 100 \qquad (16)$$

where N is the quantity being compared. The results from the simplified case can be seen in Figure 7.

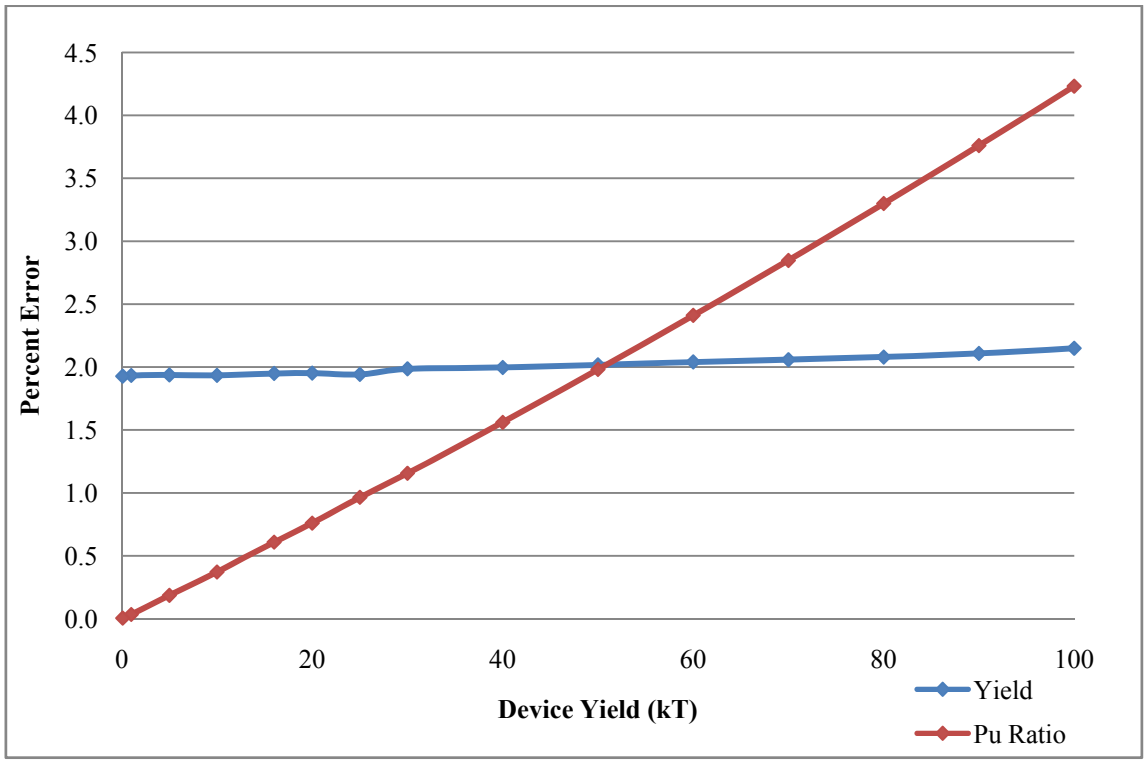

Figure 7: Percent Error in $^{240}Pu/^{239}Pu$ Ratio and Burn-up for the Simplified Case

The error in the calculated burn-up is approximately two percent for all yields between 0.1 kT and 100 kT. The error in the initial Plutonium isotopic ratio increases linearly with yield of the device. The plutonium ratio reaches two percent error at a yield of 50 kT. The error in the plutonium isotopic ratio stays well below two percent between 0.1 kT and 30 kT. This is the expected range of an improvised nuclear device. So these results show that this method can produce reasonably accurate results in the yield range for IND's.

Realistic Cases

In testing the methodology on the realistic case, yields of 10, 20, and 30 kT were used. The system performed very well. As expected, with an increase in the burn up, the errors in the higher plutonium isotopes increased. The largest errors were seen in the ^{242}Pu and ^{241}Am ratios. The largest of these errors was just over six percent. It is noteworthy that the change from the simplified case to the realistic case did not significantly change the amount of error in the ^{240}Pu/^{239}Pu ratio and calculated yield. The results of the three tests are graphed by calculated quantity together in Figure 8. The data for each of the three simulations can be seen in Table 6, Table 7, and Table 8.

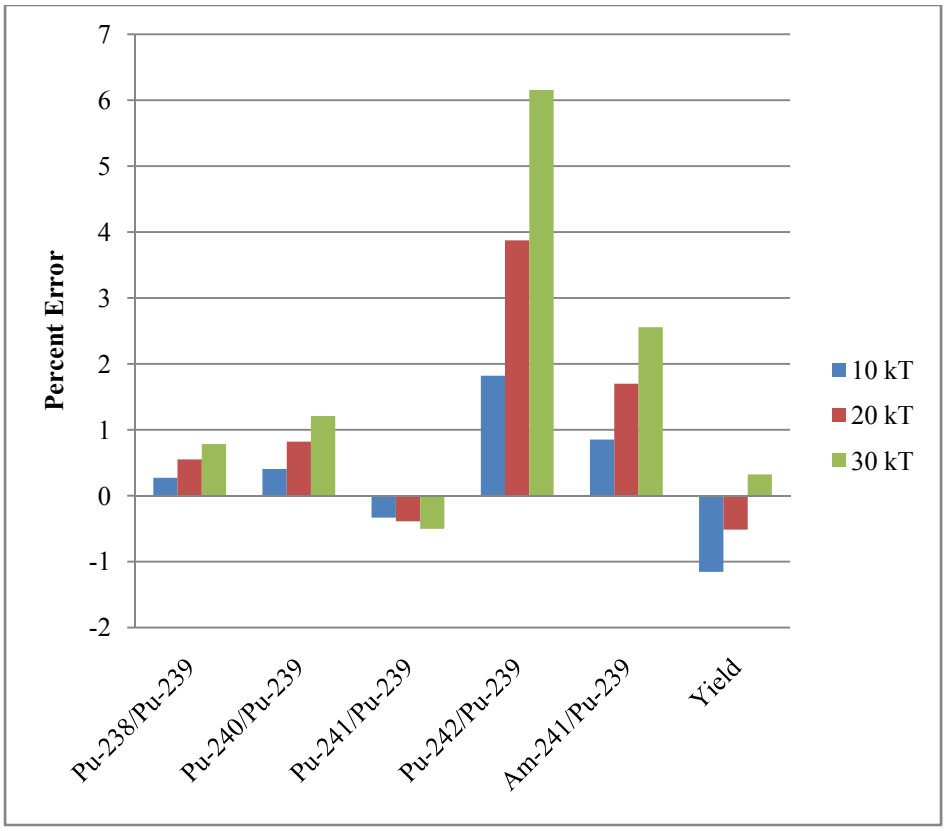

Figure 8: Weapons Grade Plutonium Simulation Results for 10, 20, and 30 kT

Table 6: Weapons Grade Pu Simulation at 10 kT Data

Nuclide Ratio	Actual (ORIGEN2)	Calculated	Percent Error
$^{238}Pu/^{239}Pu$	5.243E-05	5.280E-05	0.27
$^{240}Pu/^{239}Pu$	5.054E-02	5.053E-02	0.41
$^{241}Pu/^{239}Pu$	1.295E-03	1.280E-03	-0.33
$^{242}Pu/^{239}Pu$	5.433E-05	5.463E-05	1.82
$^{241}Am/^{239}Pu$	9.370E-04	9.372E-04	0.85
Burn-up (MWd/MT)	6.053E+03	5.983E+03	-1.16

Table 7: Weapons Grade Pu Simulation at 20 kT Data

Nuclide Ratio	Actual (ORIGEN2)	Calculated	Percent Error
$^{238}Pu/^{239}Pu$	5.265E-05	5.294E-05	0.55
$^{240}Pu/^{239}Pu$	5.033E-02	5.074E-02	0.82
$^{241}Pu/^{239}Pu$	1.284E-03	1.279E-03	-0.39
$^{242}Pu/^{239}Pu$	5.365E-05	5.573E-05	3.87
$^{241}Am/^{239}Pu$	9.292E-04	9.450E-04	1.70
Burn-up (MWd/MT)	1.211E+04	1.204E+04	-0.51

Table 8: Weapons Grade Pu Simulation at 30 kT Data

Nuclide Ratio	Actual (ORIGEN2)	Calculated	Percent Error
$^{238}Pu/^{239}Pu$	5.265E-05	5.307E-05	0.78
$^{240}Pu/^{239}Pu$	5.033E-02	5.093E-02	1.21
$^{241}Pu/^{239}Pu$	1.284E-03	1.278E-03	-0.50
$^{242}Pu/^{239}Pu$	5.365E-05	5.695E-05	6.15
$^{241}Am/^{239}Pu$	9.292E-04	9.530E-04	2.56
Burn-up (MWd/MT)	1.816E+04	1.822E+04	0.32

Similarly, the Reactor Grade plutonium case was tested using a series of yields: 10 kT, 20 kT, and 30 kT. The results of the Reactor Grade simulation are similar, but

mirror images of the Weapons Grade simulation. The results of the three simulations can be seen in Figure 9. The data from each test is located in Table 9, Table 10, and Table 11 respectively. The absolute values for each error increase in the same pattern as the previous sample simulations. However, the largest difference is that the methodology under predicts the ratio for each isotope except ^{241}Am. This is due to the assumptions used in deriving the methodology. Since each of the minor (non ^{239}Pu) isotopes have higher atom densities in the material, neglecting secondary effects on these isotopes introduces additional error than before.

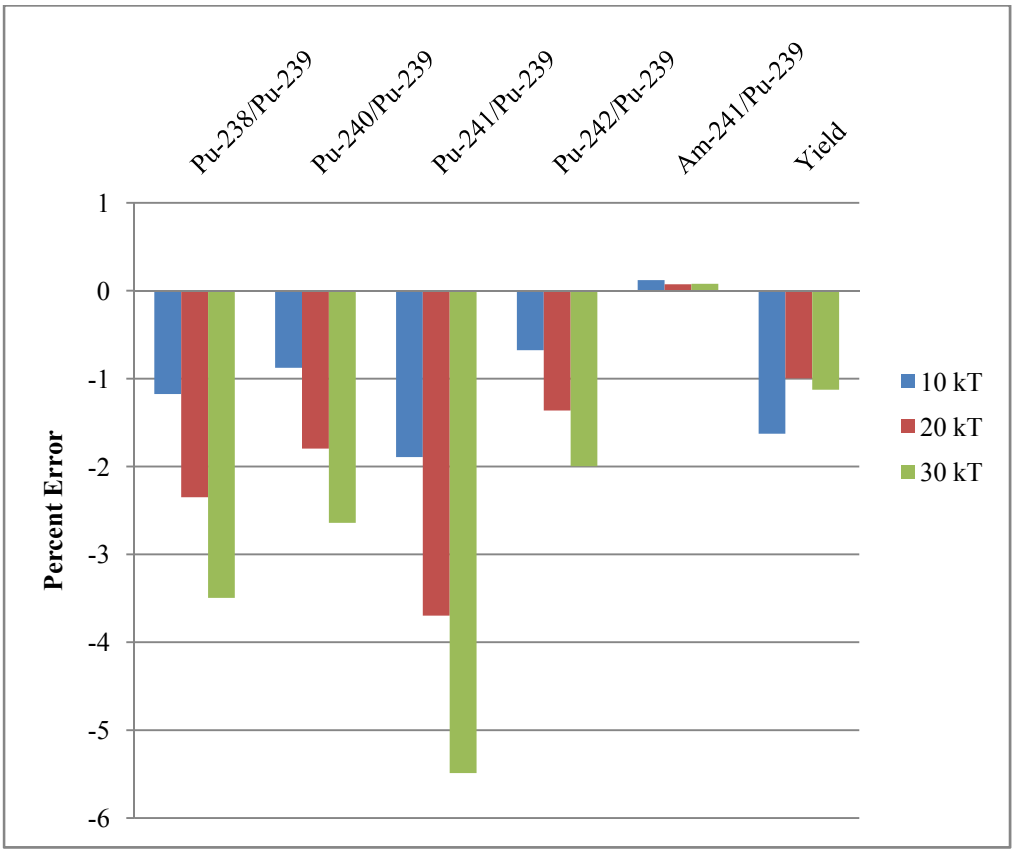

Figure 9: Reactor Grade Pu Simulation Results for 10, 20, and 30 kT

Table 9: Reactor Grade Pu Simulation at 10 kT Data

Nuclide Ratio	Actual (ORIGEN2)	Calculated	Percent Error
$^{238}Pu/^{239}Pu$	5.308E-02	5.245E-02	-1.17
$^{240}Pu/^{239}Pu$	4.713E-01	4.672E-01	-0.88
$^{241}Pu/^{239}Pu$	1.288E-01	1.263E-01	-1.89
$^{242}Pu/^{239}Pu$	1.222E-01	1.214E-01	-0.68
$^{241}Am/^{239}Pu$	6.027E-02	6.034E-02	0.12
Burn-up (MWd/MT)	6.053E+03	5.955E+03	-1.63

Table 10: Reactor Grade Pu Simulation at 20 kT Data

Nuclide Ratio	Actual (ORIGEN2)	Calculated	Percent Error
$^{238}Pu/^{239}Pu$	5.308E-02	5.183E-02	-2.35
$^{240}Pu/^{239}Pu$	4.713E-01	4.629E-01	-1.80
$^{241}Pu/^{239}Pu$	1.288E-01	1.240E-01	-3.70
$^{242}Pu/^{239}Pu$	1.222E-01	1.206E-01	-1.36
$^{241}Am/^{239}Pu$	6.027E-02	6.031E-02	0.07
Burn-up (MWd/MT)	1.211E+04	1.199E+04	-1.00

Table 11: Reactor Grade Pu Simulation at 30 kT Data

Nuclide Ratio	Actual (ORIGEN2)	Calculated	Percent Error
$^{238}Pu/^{239}Pu$	5.308E-02	5.122E-02	-3.49
$^{240}Pu/^{239}Pu$	4.713E-01	4.589E-01	-2.64
$^{241}Pu/^{239}Pu$	1.288E-01	1.217E-01	-5.49
$^{242}Pu/^{239}Pu$	1.222E-01	1.198E-01	-2.00
$^{241}Am/^{239}Pu$	6.027E-02	6.032E-02	0.08
Burn-up (MWd/MT)	1.816E+04	1.796E+04	-1.13

An interesting trend is seen in Figure 8 and Figure 9. By examining each isotope individually over the three different yields, the increase in error for each step is approximately the amount of error in the 10 kT simulation. This is constant through all

of the isotopic ratios. From this result, the error in each term is correlated to the amount of flux simulated. Potentially this relationship could be used to reduce or predict the expected error in a specific ratio.

Sensitivity Analysis

Twelve physical constants are used in the system of equations for the inverse method. These constants consist of fission yields, cross-sections, masses, and the energy recoverable from fission. Errors and uncertainties in these constants could have a significant impact on the final results. Typically, some of the cross-section data for plutonium is known very well due to years of weapons testing and design. The uncertainty in this particular quantity will come from choosing which spectrum averaged cross-sections to use. The cross-sections built in to the ORIGEN2 libraries were used here. If there were a need to use this methodology, the data from a weapons code would likely be used. The ORIGEN2 library FFTF values are very similar to the fission spectrum averages. Likewise, each of the fission yields, energy recoverable per fission, and initial plutonium atomic mass could contribute to errors in the final results. The input data from a mass spectrometry analysis should be relatively accurate and the bulk of the error and uncertainty will be introduced by the assumptions made about the device.

Equivalent Uncertainty

Several of the physical constants are used multiple times in the inverse methodology. The reoccurring constants are expected to have the greatest impact on the final output of the system. In order to test the relative effects of each individual

constant, each was perturbed independently of the others by an increase of five percent. The weapons grade plutonium sample was the baseline for these perturbations. These perturbations were run at a yield of 10 kT.

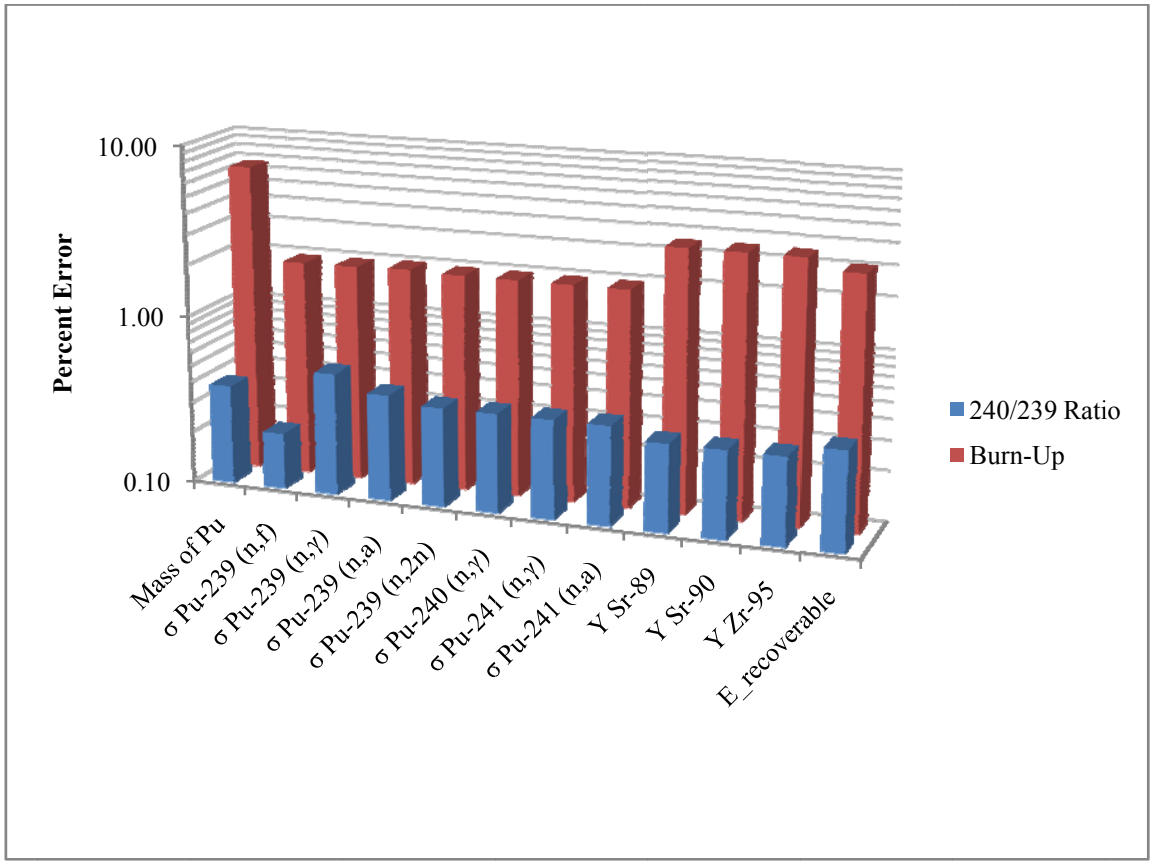

Figure 10: Errors Caused by Five Percent Perturbations in Physical Constants

As expected, each constant only affected specific ratios. Figure 10 shows the largest change seen by perturbing each variable. The fission yield constant increases affected every answer, but again these changes were all very small. The largest change was in the calculated yield. The initial Pu mass and recoverable energy had a significant

impact on the yield calculation, but did not affect the ratios at all. This effect is expected since these quantities essentially are only introduced to find the burn-up.

Orthogonal Random Sampling Method

As can be seen in Chapter III, the inverse methodology is complex. As a result, the uncertainty propagation is equally complex. This complexity makes traditional error propagation through the equations impractical. In order to study the sensitivity of the methodology, an orthogonal random sampling was performed instead. This technique allows for multiple perturbations of the constants simultaneously.

Some of the uncertainties were particularly hard to find. One instance was the uncertainty in the fission yield data. Using raw data from the Los Alamos National Laboratory data archive known as T-2 [44], a percent uncertainty was found for each of the fission yields for the fission of ^{239}Pu. Each fission product has several values. These were combined using a weighted average. This was then applied to the fission yield being applied in the methodology. Other uncertainties were found in recent journal articles and the libraries of the ORIGEN2 code. [45] [38] The uncertainties used for each variable can be found in Table 12.

Each of the variables containing uncertainty is assumed to have a normally distributed error function. This is a fairly good approximation due to the tendency of processes in physics to follow the normal distribution as described in the central limit theorem. [46] For each variable, the uncertainty was expanded to a three-sigma spread. Then using a random number generator a value within the range was selected for each constant. For this analysis, one variable was changed. For each subsequent test, the

previously randomly sampled variables and one additional variable was also selected to be changed. These constants were then fed into the methodology to observe changes in the output of the system. The perturbations for this sampling can be seen in Figure 11. The table shows that even with significant changes in the constants, only one time out of twelve did the results differ significantly from the calculations done using the standard values. The results from the orthogonal sampling can be seen in Figure 12. The data for this analysis is contained in APPENDIX C. As a result, the methodology is not overly sensitive to normal uncertainty perturbations in the system.

Table 12: Uncertainties of Constants Used in Orthogonal Random Sampling

Variable	Percent Uncertainty
Mass of Pu	1.00
σ Pu-239 (n,f)	6.50
σ Pu-239 (n,γ)	13.00
σ Pu-239 (n,a)	5.00
σ Pu-239 (n,2n)	25.00
σ Pu-240 (n,γ)	13.38
σ Pu-241 (n,γ)	3.00
σ Pu-241 (n,a)	5.00
Y Sr-89	3.69
Y Sr-90	4.26
Y Zr-95	11.63
E recoverable	5.00

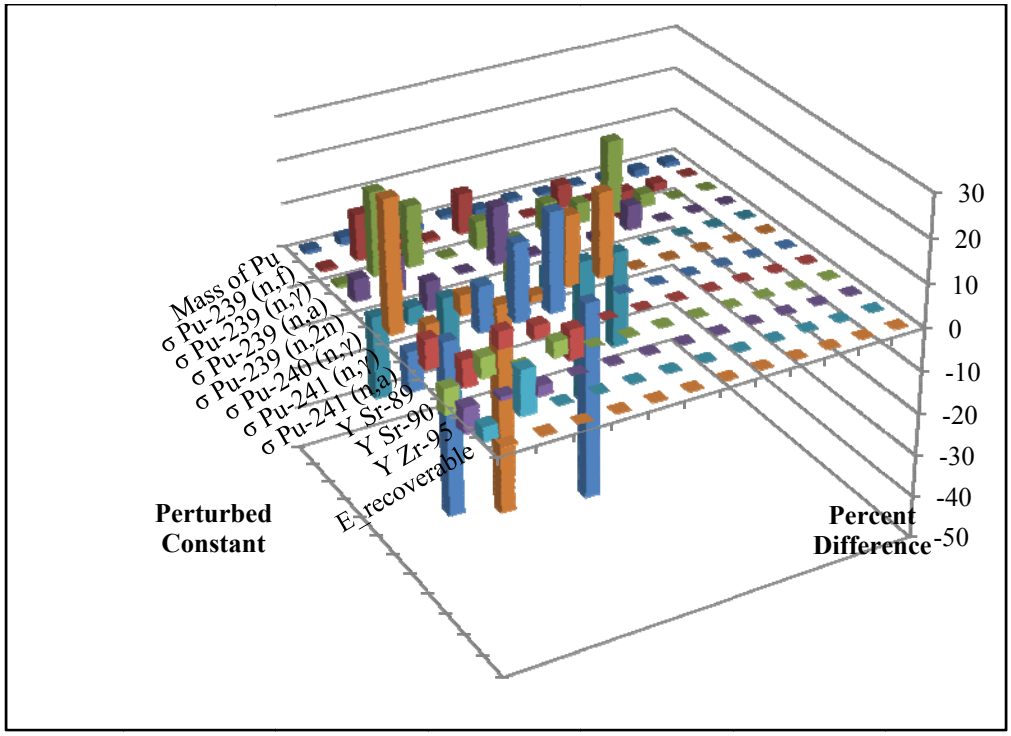

Figure 11: Randomly Sampled Perturbations of Physical Constants

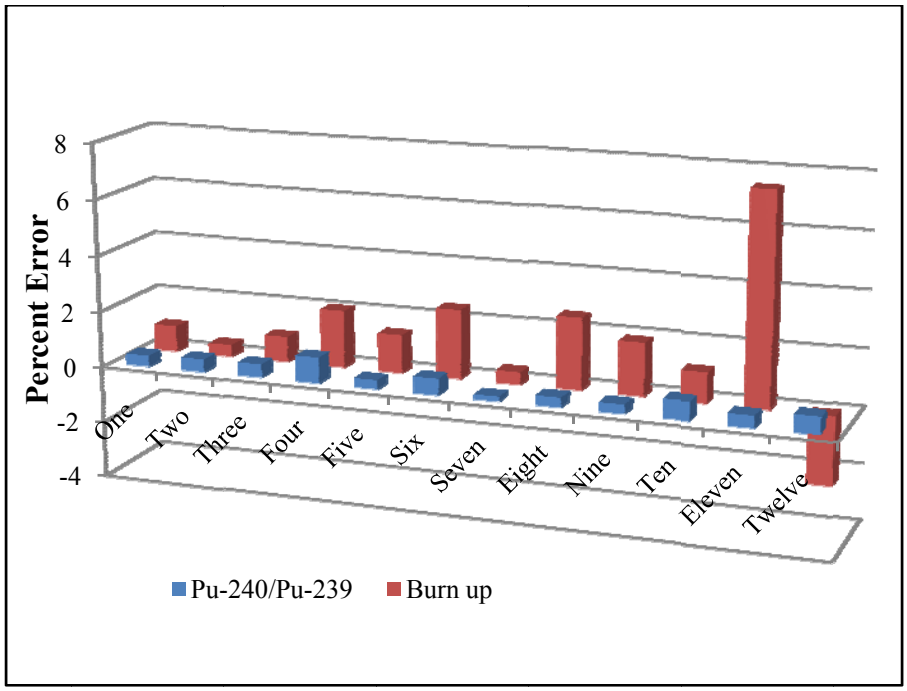

Figure 12: Orthogonal Random Sampling Results

CHAPTER V

CONCLUSIONS

A computationally efficient model that could use post-detonation environmental data to calculate pre-detonation isotopic ratios for the plutonium comprising a terrorist improvised nuclear device was developed and tested. This work completed independently of any classified work being done at the national laboratory level.

This methodology was developed and implemented in Microsoft Excel. Due to its simplicity of operation, it can be run on any computer that can handle the Microsoft Office software. This methodology explores the concept of post-event attribution in a way that incorporates standard nuclear engineering assumptions made in reactor calculations to provide insight into the origins of a nuclear weapon. The method is based on the first principles of neutron interactions to attack the inverse problem from basic fundamentals. This inverse methodology was tested against possible scenarios.

This testing showed that the calculated values of this methodology are reasonably accurate. This work sacrifices some accuracy for speed of computation; however, the results of the calculation are within two percent of the expected values for all but the ^{242}Pu ratios. As the amount of energy released in the detonation increases, the uncertainty increases in the calculations. This uncertainty is a result of the number of increased fission in isotopes other than ^{239}Pu. This increase in error is linear and should be predictable and thus correctable in practical use.

Since this work examined a very specific IND design, several limitations arise for applying the methodology to a wider spectrum of devices. This program would not

be appropriate for analysis of data from a boosted weapon. This work does not consider uranium based weapons. For uranium based IND's, a separate system would be used similar to one described in the previous works section of Chapter I.

The output of this methodology has very good potential to be used as input to another more computationally intensive and sophisticated method. Ideally the output from this work should narrow the input parameters and provide a better initial guess at the solution. Further, while another method is being run, these results could potentially rule out several sources of plutonium. In the nuclear forensics world, quickly ruling out possible origins is nearly as important as knowing the exact origin.

Several endeavors can be pursed to improve upon the methodology presented in this work.

(1) This methodology could be tested using a different and more weapons specific forward model to better assess its viability.

(2) A multi-group approach could be considered to increase the accuracy of the fission product yields, cross sections, and energies released.

(3) An iterative approach could be used in conjunction with a forward model to determine characteristics about the design of the weapon.

(4) This work could be coupled to a more sophisticated attribution effort to be tested as initial input data.

(5) Exotic materials and contaminants should be considered. This would include testing potential spoofing techniques as well.

The work presented here is a first principles based development of a post-event attribution methodology. Efforts have been made to ensure that the methodology is geometry independent in order to reduce the number of embedded assumptions. The result was a reasonably accurate system that has the potential to be used as an input mechanism for a more sophisticated attribution system. This work has the potential to significantly decrease the overall time required to complete the attribution process in the event of a nuclear IND detonation.

REFERENCES

1. J. Fox, "D.P.R.K Test Spurs US to Discuss Nuclear Attribution." *Global Security Newswire.* (October 20, 2006)

2. W.J. Broad, "Addressing the Unthinkable, US Revives Study of Fallout." *New York Times.* (March 19, 2004)

3. K. McCloud and M. Osborne, "WMD Terrorism and Usama Bin Laden." Center for Nonproliferation Studies. [Online] November 20, 2001. [Cited: September 30, 2007.] http://cns.miis.edu/pubs/reports/binladen.htm.

4. Jane's Information Group. Nuclear Proliferation (Pakistan). *Jane's Chemical, Biological, Radiological, and Nuclear Assesments.* [Online] August 4, 2008. [Cited: September 30, 2008] http://www.janes.com/extracts/extract/jcbrn/jcbrna440.html.

5. D. Albright and P. Brannan, "The Al Kibar Reactor: Extraordinary Camouflage, Troubling Implications." [Online] May 12, 2008. [Cited: September 30, 2008] http://www.isis-online.org/publications/syria/SyriaReactorReport_12May2008.pdf.

6. E. MacAskill, "U.S. Claims North Korea Helped Build Syria Reactor Plant." *The Guardian (UK).* (April 25, 2008)

7. J. Doyle, *Nuclear Safeguards, Security, and Nonproliferation: Achieving Security with Technology and Policy.* Butterworth-Heinemann: Burlington, MA. (2008) ISBN: 978-0-7506-8673-0.

8. J. Holdren and M. Bunn, "Nuclear Basics." *Securing the Bomb: Blocking the Terrorist Pathway to the Bomb.* [Online] November 25, 2005. [Cited: September 30, 2008.] http://www.nti.org/e_research/cnwm/overview/technical1.asp.

9. R. Serber, *Los Alamos Primer.* Los Alamos National Laboratory: Los Alamos, NM. (April 1943)

10. C. Loeber, *Building the Bombs: A History of the Nuclear Weapons Complex.* Sandia National Laboratories: Albuquerque, NM (2002)

11. M. Miller, "Nuclear Attribution as Deterrence." *Nonproliferation Review.* **14**, *1*. pp 33-60. (2007)

12. J. Davis, "The Attribution of WMD Events." *Homeland Security Journal.* (April 2003)

13. A.C. Hayes and G. Jungman, "Domestic Nuclear Event Attribution: Proliferant Devices Based on Reactor-Grade Fuel." Theoretical Division Nuclear Weapons Program Highlights. Los Alamos National Laboratory: Los Alamos, NM (2005-2006)

14. Table of Nuclides. *Nuclear Data Evaluation Laboratory.* [Online] Korea Atomic Energy Research Institute, 2000. [Cited: September 30, 2008.] http://atom.kaeri.re.kr/.

15. M.J. Kristo, D.K. Smith, S. Neimeyer, G.B. Dudder. "Model Action Plan for Nuclear Forensics and Nuclear Attribution: A Draft Product for the International Technical Working Group". Lawrence Livermore National Laboratory: Livermore, CA. (March 3, 2004)

16. W. Dunlop and H. Smith, "Who Did It? Using International Forensics to Detect and Deter Nuclear Terrorism." *Arms Control Today.* [Online] (October 2006). [Cited: Feb 13, 2007.]

17. C-ACS Forensics and Attribution. *Chemistry Division: Los Alamos National Laboratory.* [Online] [Cited: June 6, 2007.] http://peral1.lanl.gov/external/ACS/char/att.htm.

18. A. Heller, "Identifying the Source of Stolen Nuclear Materials." *Science & Technology Review.* (Jan/Feb, 2007)

19. I. Hutcheon, "Tracing the Steps in Nuclear Material Trafficking." *Science & Technology Review.* (March, 2005)

20. K. Moody, I. Hutcheon, and P. Grant. *Nuclear Forensic Analysis.* Taylor & Francis, CRC: Boca Raton, (2005). ISBN: 0-8493-1513-1.

21. W.B. Doriese, J.N. Ullom, J.A. Beall, W.D. Duncan, L. Ferreira et al. "14-Pixel, Multiplexed Array of Gamma-Ray Microcalorimeters with 47 eV Energy Resolution at 103 keV." *Applied Physics Letters.* **90**, 193508-1 - 193508-3(2007)

22. S.F. Terracol, S. Ali, T.R. Niedermayer, I.D. Hau, O.B. Drury et al. "Ultra-High Resolution Gamma-Ray Spectrometer Development for Nuclear Attribution and Non-Proliferation Applications." *Nuclear Science Symposium Conference Record.* IEEE, (2004)

23. M. Scott, "Nuclear Forensics: Attributing the Source of Spent Fuel Used in an RDD Event." Masters Thesis, Texas A&M University: College Station, TX (2005)

24. A. LaFleur, "Advancing Methods for Determining the Source of HEU Used in a Terrorist Nuclear Weapon." Undergraduate Honors Thesis, Texas A&M University: College Station, TX (2007)

25. M. Wallenius, P. Peerani, and L. Koch. "Origin Determination of Plutonium Materials in Nuclear Forensics." *Journal of Radioanalytical and Nuclear Chemistry.* **246**, *2*, 317-321. (2000)

26. M. Wallenius, A. Morgenstern, C. Apostolidis, K. Mayer. "Determination of the Age of Highly Enriched Uranium." *Analytical and Bioanalytical Chemistry.* **374**, *3*, 379-384. (2002)

27. M. Wallenius, K. Mayer. "Age Determination of Plutonium Material in Nuclear Forensics by Thermal Ionization Mass Spectrometry." *Journal of Analytical Chemistry.* **366**, *3*, 234-238. (2000)

28. M. Wallenius. "Origin Determination of Reactor Produced Plutonium by Mass Spectrometric Techniques: Application to Nuclear Science and Safeguards." Ph.D. Dissertation. University of Helsinki: Helskini, Finland, (2001)

29. K. Mayer. "Nuclear Forensics - A Methodology Providing Clues on the Origin of Illicitly Trafficked Nuclear Materials." *The Analyst.* **130**, 433-441 (2005)

30. *Nuclear Forensics: Role, State of the Art, Program Needs.* Joint Working Group of the American Physical Society and the American Association for the Advancement of Science.

31. H. Aigner, R. Binner, E. Kuhn, U. Blohm-Hieber, K. Mayer, et al. "International Target Values 2000 for Measurement Uncertanties in Safeguarding Nuclear Materials." [Online] 2000. [Cited: September 30, 2008.] http://www.inmm.org/publications/JNMMPaperITV.pdf.

32. A. DeVolpi, "Fissle Materials and Nuclear Weapons Proliferation." *Ann. Rev. Nucl. Part. Sci.* **36**, 83-114. (1986)

33. L. Bloomfield. "Nuclear Weapons." *How Things Work.* [Online] August 18, 2008. [Cited: September 30, 2008.] http://howthingswork.virginia.edu/nuclear_weapons.html.

34. X-5 Monte Carlo Team. *MCNP - A General Monte Carlo N-Particle Transport Code, Version 5.* Los Alamos National Laboratory: Los Alamos, NM. LA-UR-03-1987. (2004)

35. TransWare Enterprises, Inc. *TransFX Computer Software Manuals.* San Jose, CA : Published Electronically, (2002) http://www.twe.com/index.php/nuclear

36. W.S. Charlton, W.D. Stanbro, R.T. Perryl. "Comparisons of Helios, ORIGEN2, and Monteburns Calculated Am-241 and Am-243 Concentrations to Measured Values for PWR, BWR, and VVER Spent Fuel." *Journal of Nuclear Science and Technology.* **37**, *7*, 615-623. (2000)

37. W.S. Charlton, R.T. Perry, B.L. Fearey, T.A. Paris. "Comparisons of HELIOS Calculated Isotope Concentrations to Measured Values for Several Reactor Systems." *Procedings of the 16th Fuel Management Systems User Group Meeting,* Madrid (ES). October 18, 1998. **16**. (1998)

38. A.G. Croff, *A User's Manual for the ORIGEN2 Computer Code.* Oak Ridge National Laboratory: Oak Ridge, TN, ORNL/TM7175. (1980)

39. A.G. Croff. "ORIGEN2: A Versitile Computer Code for Calculating the Nuclide Compositions and Characteristics of Nuclear Materials." *Nuclear Technology.* **62**, 335-352. (1983)

40. J.C. Miller and W.S. Charlton. "Analytical Inverse Model for the Post-Event Attribution of Plutonium." *Proceedings of the Annual Meeting of the Institute of Nuclear Materials Management.* 48th Meeting, July 2007, Tucson, AZ. Institute of Nuclear Materials Management (2007)

41. International Atomic Energy Agency. Nuclear Data Section. *Nuclear Data for Safeguards.* [Online] 2004. [Cited: September 30, 2008.] http://www-nds.iaea.org/sgnucdat/c1.htm.

42. Lockheed Martin. *Nuclides and Isotopes: Chart of the Nuclides.*16th Edition.: Knolls Atomic Power Laboratory: Niskayuna, NY (2002).

43. J.C. Mark, "Explosive Properties of Reactor-Grade Plutonium." *Science & Global Security.* **4**, 111-128. (1993)

44. T.R. England and B.F. Rider. "Evaluation and Compilation of Fission Product Yields." *Evaluated Nuclear Data Files -349.* (1993)

45. G. Aliberti, G. Palmiotti, M. Salvatores, C.G. Stenberg. "Impact of Nuclear Data Uncertainties on Transmutation of Actinides in Accelerator-Driven Assemblies." *Nuclear Science and Engineering.* **146**, *1*,13-50. (2004)

46. E. Weisstein. "Normal Distribution." *MathWorld - A Wolfram Web Resource.* [Online] [Cited: September 30, 2008.] http://mathworld.wolfram.com/NormalDistribution.html.

APPENDIX A

EXAMPLE ORIGEN2 INPUT DECK

ORIGEN2 Deck for a 10 kT Blast of Weapons Grade Sample

```
-1
-1
-1
 RDA PU sphere - swab 4
 RDA 1 MT Pu
 LIB  0 1 2 3 381 382 383 9 50 0 1 0
 INP  1   1  -1  -1   1   1
 BUP
 IRP  6000.0 87166.665   1 2 1 2 BURNUP
 DEC  1.0          2 3 4 0 DECAY= 1 day after blast
 DEC  7.0          3 4 4 0 DECAY= 7 days after blast
 BUP
 OPTL 2*8 8 8 8 23*8
 OPTA 2*8 8 8 5 14*8 5 8 5 6*8
 OPTF 2*8 8 8 5 23*8
 OUT  4   1  -1   0
 END
2 922340 4.6    942380 49.8    942390 949800  942400  48000
2 942410 1230   942420 51.6    952410 890.0   932370  8.8
 0
```

APPENDIX B

COMPLETE INVERSE METHODOLOGY DERIVATION

Time of Explosion:

Based on the small yield fraction of the Yttrium nuclides, we will use concentrations of Yttrium to determine the exact time of the detonation. This seems trivial because it is pretty obvious when a nuclear weapon is detonated; however, this determination will date the precise time of detonation in order to correct for decay. Also, by knowing the exact amount of decay time, we can confirm the time of detonation as a check to make sure that our equations and program is functioning properly.

First we recall the basic decay equations for parent and daughter nuclides:

$$N_{Sr-89}(t) = N_{Sr-89}(0)e^{-\lambda_{Sr-89}t} \quad (1)$$

$$N_{Y-89}(t) = N_{Sr-89}(0)\left[1 - e^{-\lambda_{Sr-89}t}\right] \quad (2)$$

These equations assume that the entire production of Yittrium-89 is from the decay of Strontium-89. This is a reasonable assumption as the fission yield for Yt-89 is quite low. Next we will divide the Sr-89 equation into the Yt-89 equation.

$$\frac{N_{Y-89}(t)}{N_{Sr-89}(t)} = \frac{N_{Sr-89}(0)}{N_{Sr-89}(0)}\frac{\left[1 - e^{-\lambda_{Sr-89}t}\right]}{e^{-\lambda_{Sr-89}t}} \quad (3)$$

$$\frac{N_{Y-89}(t)}{N_{Sr-89}(t)} = e^{-\lambda_{Sr-89}t} - 1 \quad (4)$$

Then with a little algebraic work we can find an expression for the decay time based on the ratio of Y to Sr at any time T.

$$T_{decay} = \frac{1}{\lambda_{Sr-89}} ln\left[\frac{N_{Y-89}(t)}{N_{Sr-89}(t)} + 1\right] \quad (5)$$

$$T_{event} = T_{measurement} - T_{decay} \quad (6)$$

In looking at this problem we will first examine the atomic densities of known fission products. The fission products that we are interested in are Strontium-89, Zirconium-95, and Strontium-90.

Strontium-90:

First, we will start with our basic change rate equation. This is the difference between the production and loss rates:

$$\frac{dN_{Sr-90}}{dt} = \{production\} - \{loss\} \tag{7}$$

$$\frac{dN_{Sr-90}}{dt} = Y_{Sr-90}\Sigma_f^{Pu-239}\varphi - \lambda_{Sr-90}N_{Sr-90} - \sigma_{Sr-90}N_{Sr-90}\varphi \tag{8}$$

Now we will simplify this equation. First, we know that the half life of Strontium-90 is approximately 28.78 years according to the Chart of Nuclides. Thus we will not need to worry about any significant decay over our desired time frame. Secondly, the cross section of Strontium-90 is dominated by elastic scattering (4.8 barns of 5.6 barns according to atom.kaeri.re.kr…). Thus our equation simplifies:

$$\frac{dN_{Sr-90}}{dt} = Y_{Sr-90}\Sigma_f^{Pu-239}\varphi \tag{9}$$

Next, we will rearrange and integrate over a time T for fission to occur during the detonation process which will be approximately 10^{-6} seconds.

$$dN_{Sr-90} = Y_{Sr-90}\Sigma_f^{Pu-239}(t)\varphi(t)dt \tag{10}$$

$$\int_0^T dN_{Sr-90} = Y_{Sr-90}\int_0^T \Sigma_f^{Pu-239}(t)\varphi(t)dt \tag{11}$$

$$N_{Sr-90} = Y_{Sr-90}\int_0^T \Sigma_f^{Pu-239}(t)\varphi(t)dt \tag{12}$$

Next we will make an approximation which will ease our calculations. The first of which is dealing with the device yield.

$$E_{yield} = V_{volume} E_{released} \int_0^T \Sigma_f^{Pu-239}(t)\varphi(t)dt \tag{13}$$

$$\frac{E_y}{VE_r} = \int_0^T \Sigma_f^{Pu-239}(t)\varphi(t)dt \tag{14}$$

We define burn up (BU) to be the yield of the device over the mass of the fissile material. Also we define the density of the Plutonium-239 in our device. Thus:

$$\Psi = \frac{E_y}{m_0^{Pu}} \tag{15}$$

$$\rho^{Pu} = \frac{m_0^{Pu}}{V} \tag{16}$$

Then combining these definitions, we have:

$$\int_0^T \Sigma_f^{Pu-239}(t)\varphi(t)dt = \Psi \frac{1}{E_r} \rho^{Pu-239} \tag{17}$$

$$N_{Sr-90} = Y_{Sr-90} \int_0^T \Sigma_f^{Pu-239}(t)\varphi(t)dt \tag{18}$$

$$N_{Sr-90} = Y_{Sr-90} \Psi \frac{1}{E_r} \rho^{Pu-239} \tag{19}$$

We are really interested in the ratio of the Strontium-90 to the Plutonium-239 left. Thus we introduce that quantity.

$$N_{Sr-90}(T) \frac{N_{Pu-239}(T)}{N_{Pu-239}(T)} = Y_{Sr-90} \Psi \frac{1}{E_r} \rho^{Pu-239} \tag{20}$$

$$\frac{N_{Sr-90}(T)}{N_{Pu-239}(T)} = Y_{Sr-90} \Psi \frac{1}{E_r} \frac{\rho^{Pu-239}}{N_{Pu-239}(T)} \tag{21}$$

$$\frac{N_{Sr-90}(T)}{N_{Pu-239}(T)} = Y_{Sr-90}\Psi\frac{1}{E_r}\frac{N_{Pu}(0)}{N_{Pu-239}(T)}\frac{m_{Pu}}{N_a}\frac{N_{Pu-239}(0)}{N_{Pu-239}(0)} \tag{22}$$

$$\frac{N_{Sr-90}(T)}{N_{Pu-239}(T)} = Y_{Sr-90}\frac{1}{E_r}\frac{m_{Pu}}{N_a}\left\{\Psi\frac{N_{Pu-239}(0)}{N_{Pu-239}(T)}\frac{N_{Pu}(0)}{N_{Pu-239}(0)}\right\} \tag{23}$$

We will now substitute a variable for the bracketed quantity as shown here.

$$\Psi_X = \left\{\Psi\frac{N_{Pu-239}(0)}{N_{Pu-239}(T)}\frac{N_{Pu}(0)}{N_{Pu-239}(0)}\right\} \tag{24}$$

Then by making this substitution, we have:

$$\frac{N_{Sr-90}(T)}{N_{Pu-239}(T)} = Y_{Sr-90}\frac{1}{E_r}\frac{m_{Pu}}{N_a}\Psi_X \tag{25}$$

Now we have an equation with known or measurable quantities, which will allow us to solve for the bracketed quantity. Likewise we can find a similar equation for Strontium-89 and Zirconium-95. The only difference between those two equations will be the quantity for the measured fission product and yield.

Plutonium-240:

Next we wish to examine Plutonium-240 with our same change rate equation:

$$\frac{dN_{Pu-240}}{dt} = \{production\} - \{loss\} \tag{26}$$

$$\frac{dN_{Pu-240}}{dt} = \Sigma_a^{Pu-239}\varphi - \lambda_{Pu-240}N_{Pu-240} - \Sigma_a^{Pu-240}\varphi \tag{27}$$

Since Pu-240 has approximately a 6564 year half life. We are going to neglect our decay term over our time period. Also, in examining the cross sections of Plutonium-240, we find that the total cross section is 7.7 barns and the scattering cross section is 5.2 barns. Thus we may also neglect this term.

$$\frac{dN_{Pu-240}}{dt} = \Sigma_a^{Pu-239}\varphi(t) \tag{28}$$

$$\frac{dN_{Pu-240}}{dt} = N^{Pu-239}\sigma_\gamma^{Pu-239}\varphi(t)\frac{\sigma_f^{Pu-239}}{\sigma_f^{Pu-239}} \tag{29}$$

$$\frac{dN_{Pu-240}}{dt} = \left[N^{Pu-239}\sigma_f^{Pu-239}\varphi(t)\right]\frac{\sigma_\gamma^{Pu-239}}{\sigma_f^{Pu-239}} \tag{30}$$

Now we will substitute in the definition of the macroscopic cross-section. Thus we get:

$$dN_{Pu-240} = \frac{\sigma_\gamma^{Pu-239}}{\sigma_f^{Pu-239}}\Sigma_f^{Pu-239}(t)\varphi(t)dt \tag{31}$$

$$\int_0^T dN_{Pu-240} = \frac{\sigma_\gamma^{Pu-239}}{\sigma_f^{Pu-239}}\int_0^T \Sigma_f^{Pu-239}(t)\varphi(t)dt \tag{32}$$

Now, we will recall our simplification of our macroscopic cross-section and flux integral from before:

$$\int_0^T \Sigma_f^{Pu-239}(t)\varphi(t)dt = \Psi \frac{1}{E_r}\rho^{Pu-239} \tag{33}$$

The resulting equation is as follows:

$$N_{Pu-240}(T) - N_{Pu-240}(0) = \frac{\sigma_\gamma^{Pu-239}}{\sigma_f^{Pu-239}}\Psi \frac{1}{E_r}\rho^{Pu-239} \tag{34}$$

$$\frac{N_{Pu-240}(T) - N_{Pu-240}(0)}{N_{Pu-239}(T)} = \frac{\sigma_\gamma^{Pu-239}}{\sigma_f^{Pu-239}}\Psi \frac{1}{E_r}\frac{\rho^{Pu}}{N_{Pu-239}(T)} \tag{35}$$

$$\frac{N_{Pu-240}(T) - N_{Pu-240}(0)}{N_{Pu-239}(T)} \tag{36}$$

$$= \frac{\sigma_\gamma^{Pu-239}}{\sigma_f^{Pu-239}}\Psi \frac{1}{E_r}\frac{\rho^{Pu}}{N_{Pu-239}(T)}\frac{N_a m_{Pu}}{N_a m_{Pu}}$$

Then by noting the definition of atom density, we can simplify further,

$$\frac{N_{Pu-240}(T) - N_{Pu-240}(0)}{N_{Pu-239}(T)} \tag{37}$$

$$= \frac{\sigma_\gamma^{Pu-239}}{\sigma_f^{Pu-239}}\frac{m_{Pu}}{N_a}\Psi \frac{1}{E_r}\frac{N_{Pu}(0)}{N_{Pu-239}(T)}\frac{N_{Pu-239}(0)}{N_{Pu-239}(0)}$$

Using the same approximations and substitutions as above:

$$\frac{N_{Pu-240}(T)}{N_{Pu-239}(T)} - \frac{N_{Pu-240}(0)}{N_{Pu-239}(T)} \tag{38}$$

$$= \frac{\sigma_\gamma^{Pu-239}}{\sigma_f^{Pu-239}}\frac{m_{Pu}}{N_a}\frac{1}{E_r}\left\{\Psi \frac{N_{Pu-239}(0)}{N_{Pu-239}(T)}\frac{N_{Pu}(0)}{N_{Pu-239}(0)}\right\}$$

$$\frac{N_{Pu-240}(0)}{N_{Pu-239}(T)} = \frac{N_{Pu-240}(T)}{N_{Pu-239}(T)} - \frac{\sigma_\gamma^{Pu-239}}{\sigma_f^{Pu-239}}\frac{m_{Pu}}{N_a}\frac{1}{E_r}\Psi_{\mathrm{X}} \tag{39}$$

Plutonium-239:

One of the two main fissile materials used to make nuclear weapons is Pu-239. If a weapon is made using this material, there will be traces left after the weapon is detonated. This trace signature is important. We will be comparing most of our other values to this amount of Pu-239 left after the explosion.

Again, we will start with our change rate equation:

$$\frac{dN_{Pu-239}}{dt} = \{production\} - \{loss\} \tag{40}$$

$$\frac{dN_{Pu-239}}{dt} = -\sigma_a^{Pu-239} N_{Pu-239}(t)\varphi(t) \tag{41}$$

We will try to match find a similar equation to our Pu-240, thus we will use the same methodology in deriving an equation based on Pu-239.

$$\frac{dN_{Pu-239}}{dt} = -\sigma_a^{Pu-239} \frac{\sigma_f^{Pu-239}}{\sigma_f^{Pu-239}} N_{Pu-239}(t)\varphi(t) \tag{42}$$

$$dN_{Pu-239} = -\frac{\sigma_a^{Pu-239}}{\sigma_f^{Pu-239}} \sigma_f^{Pu-239} N_{Pu-239}(t)\varphi(t)dt \tag{43}$$

$$\int_0^T dN_{Pu-239} = -\frac{\sigma_a^{Pu-239}}{\sigma_f^{Pu-239}} \int_0^T \Sigma_f^{Pu-239}(t)\varphi(t)dt \tag{44}$$

We will now insert our burn-up relation to be consistent with our equations to develop our system of equations.

$$\int_0^T dN_{Pu-239} = -\frac{\sigma_a^{Pu-239}}{\sigma_f^{Pu-239}} \Psi \frac{1}{E_r} \rho^{Pu} \tag{45}$$

$$N_{Pu-239}(T) - N_{Pu-239}(0) = -\frac{\sigma_a^{Pu-239}}{\sigma_f^{Pu-239}} \Psi \frac{1}{E_r} \rho^{Pu} \tag{46}$$

We need to get an equation of the same form as before, so we will manipulate our terms as follows:

$$\frac{N_{Pu-239}(T) - N_{Pu-239}(0)}{N_{Pu-239}(T)} = -\frac{\sigma_a^{Pu-239}}{\sigma_f^{Pu-239}} \Psi \frac{1}{E_r} \frac{\rho^{Pu}}{N_{Pu-239}(T)} \tag{47}$$

$$1 - \frac{N_{Pu-239}(0)}{N_{Pu-239}(T)} = -\frac{\sigma_a^{Pu-239}}{\sigma_f^{Pu-239}} \Psi \frac{1}{E_r} \frac{N_{Pu}(0)}{N_{Pu-239}(T)} \frac{m_{Pu}}{N_a} \tag{48}$$

$$1 - \frac{N_{Pu-239}(0)}{N_{Pu-239}(T)} = -\frac{\sigma_a^{Pu-239}}{\sigma_f^{Pu-239}} \Psi \frac{1}{E_r} \frac{m_{Pu}}{N_a} \frac{N_{Pu}(0)}{N_{Pu-239}(T)} \frac{N_{Pu-239}(0)}{N_{Pu-239}(0)} \tag{49}$$

$$1 - \frac{N_{Pu-239}(0)}{N_{Pu-239}(T)} = -\frac{\sigma_a^{Pu-239}}{\sigma_f^{Pu-239}} \frac{1}{E_r} \frac{m_{Pu}}{N_a} \left\{ \Psi \frac{N_{Pu}(0)}{N_{Pu-239}(T)} \frac{N_{Pu-239}(0)}{N_{Pu-239}(0)} \right\} \tag{50}$$

$$\frac{N_{Pu-239}(0)}{N_{Pu-239}(T)} = 1 + \frac{\sigma_a^{Pu-239}}{\sigma_f^{Pu-239}} \frac{1}{E_r} \frac{m_{Pu}}{N_a} \Psi_X \tag{51}$$

Plutonium-238:

Plutonium-238 is one of the main pollutants in the plutonium device and we will derive the relationship for this isotope's concentration before the event. Once again we will start with the basic change rate equation. We also assume that there is only production of Pu-238 by the (n,2n) reaction in Pu-239.

$$\frac{dN_{Pu-238}}{dt} = \{production\} - \{loss\} \tag{52}$$

$$\frac{dN_{Pu-238}}{dt} = \sigma_{n,2n}^{Pu-239} N_{Pu-239}(t)\varphi(t) - 0 \tag{53}$$

Similarly to our previous equations we will assume that the amount of Plutonium-239 atoms is approximately constant over the irradiation period. We will also assume a constant flux to simplify our math.

$$dN_{Pu-238} = \frac{\sigma_{n,2n}^{Pu-239}}{\sigma_f^{Pu-239}} \sigma_f^{Pu-239} N_{Pu-239}(t)\varphi(t)dt \tag{54}$$

$$\int_0^T dN_{Pu-238} = \frac{\sigma_{n,2n}^{Pu-239}}{\sigma_f^{Pu-239}} \int_0^T \Sigma_f^{Pu-239}(t)\varphi(t)dt \tag{55}$$

Using the same burn up approximation that we made earlier, we find that our equation becomes:

$$N_{Pu-238}(T) - N_{Pu-238}(0) = \frac{\sigma_{n,2n}^{Pu-239}}{\sigma_f^{Pu-239}} \Psi \frac{1}{E_r} \rho^{Pu} \tag{56}$$

Then utilizing the same substitutions as the previous derivations we see that we find a corresponding equation for Pu-238.

$$\frac{N_{Pu-238}(T) - N_{Pu-238}(0)}{N_{Pu-239}(T)} = \frac{\sigma_{n,2n}^{Pu-239}}{\sigma_f^{Pu-239}} \Psi \frac{1}{E_r} \frac{\rho^{Pu}}{N_{Pu-239}(T)} \tag{57}$$

$$\frac{N_{Pu-238}(T)}{N_{Pu-239}(T)} - \frac{N_{Pu-238}(0)}{N_{Pu-239}(T)} = \frac{\sigma_{n,2n}^{Pu-239}}{\sigma_f^{Pu-239}} \frac{1}{E_r} \frac{m_{Pu}}{N_a} \left\{ \Psi \frac{N_{Pu-239}(0)}{N_{Pu-239}(T)} \frac{N_{Pu}(0)}{N_{Pu-239}(0)} \right\} \tag{58}$$

$$\frac{N_{Pu-238}(0)}{N_{Pu-239}(T)} = \frac{N_{Pu-238}(T)}{N_{Pu-239}(T)} - \frac{\sigma_{n,2n}^{Pu-239}}{\sigma_f^{Pu-239}} \frac{1}{E_r} \frac{m_{Pu}}{N_a} \Psi_X \tag{59}$$

Plutonium-241:

Plutonium-241 is another "pollutant" in the plutonium metal of the core. This is an important piece of the isotopic concentrations of the initial device in the origin determination. Once again we will start with our basic change rate equation. We assume that change in concentrations is dominated by the (n,gamma) reaction in Pu-240. Thus we will neglect the burn up and decay of the Pu-241 (14.4 year half life).

$$\frac{dN_{Pu-241}}{dt} = \{production\} - \{loss\} \tag{60}$$

$$\frac{dN_{Pu-241}}{dt} = \sigma_{\gamma}^{Pu-240} N_{Pu-240}(t)\varphi(t) - 0 \tag{61}$$

We need to relate this isotopes production to the burn up we defined earlier. This will allow us to use a similar methodology.

$$\frac{dN_{Pu-241}}{dt} = \frac{\sigma_{\gamma}^{Pu-240}}{\sigma_{f}^{Pu-239}} \frac{N_{Pu-240}(t)}{N_{Pu-239}(t)} \sigma_{f}^{Pu-239} N_{Pu-239}(t)\varphi(t) \tag{62}$$

$$\int_0^T dN_{Pu-241} = \frac{\sigma_{\gamma}^{Pu-240}}{\sigma_{f}^{Pu-239}} \frac{N_{Pu-240}(t)}{N_{Pu-239}(t)} \int_0^T \Sigma_{f}^{Pu-239}(t)\varphi(t)dt \tag{63}$$

$$\int_0^T dN_{Pu-241} = \frac{\sigma_{\gamma}^{Pu-240}}{\sigma_{f}^{Pu-239}} \frac{N_{Pu-240}(t)}{N_{Pu-239}(t)} \Psi \frac{1}{E_r} \rho^{Pu} \tag{64}$$

We will assume that the Pu-240 to Pu-239 ratio does not change significantly over time. Thus we are able to use the value calculated before in that place. This should under predict the value of the Pu-241 ratio since it does not take into consideration the production of Pu-240 which could become Pu-241. We will also substitute our definition of burn up to get:

$$N_{Pu-241}(T) - N_{Pu-241}(0) = \frac{\sigma_\gamma^{Pu-240}}{\sigma_f^{Pu-239}} \frac{N_{Pu-240}(0)}{N_{Pu-239}(T)} \Psi \frac{1}{E_r} \rho^{Pu} \qquad (65)$$

Then we will multiply by a constant in order to find the consistent factor for the burn up.

$$\frac{N_{Pu-241}(T) - N_{Pu-241}(0)}{N_{Pu-239}(T)} = \frac{\sigma_\gamma^{Pu-240}}{\sigma_f^{Pu-239}} \frac{N_{Pu-240}(0)}{N_{Pu-239}(T)} \Psi \frac{1}{E_r} \frac{\rho^{Pu}}{N_{Pu-239}(T)} \qquad (66)$$

Then with a little algebra we are able to find the following equation.

$$\frac{N_{Pu-241}(0)}{N_{Pu-239}(T)} = \frac{N_{Pu-241}(T)}{N_{Pu-239}(T)} - \frac{\sigma_\gamma^{Pu-240}}{\sigma_f^{Pu-239}} \frac{N_{Pu-240}(0)}{N_{Pu-239}(T)} \frac{1}{E_r} \frac{m_{Pu}}{N_a} \left\{ \Psi \frac{N_{Pu-239}(0)}{N_{Pu-239}(T)} \frac{N_{Pu}(0)}{N_{Pu-239}(0)} \right\} \qquad (67)$$

$$\frac{N_{Pu-241}(0)}{N_{Pu-239}(T)} = \frac{N_{Pu-241}(T)}{N_{Pu-239}(T)} - \frac{\sigma_\gamma^{Pu-240}}{\sigma_f^{Pu-239}} \frac{N_{Pu-240}(0)}{N_{Pu-239}(T)} \frac{1}{E_r} \frac{m_{Pu}}{N_a} \Psi_X \qquad (68)$$

Plutonium-242:

Plutonium-242 is another isotope contained in the pit. This is an important piece of the isotopic concentrations of the initial device in the origin determination. Once again we will start with our basic change rate equation. We assume that change in concentrations is dominated by the (n,gamma) reaction in Pu-241. Thus we will neglect the burn up and decay of the Pu-242 (373300 year half life).

$$\frac{dN_{Pu-242}}{dt} = \{production\} - \{loss\} \tag{69}$$

$$\frac{dN_{Pu-242}}{dt} = \sigma_{\gamma}^{Pu-241} N_{Pu-241}(t)\varphi(t) - 0 \tag{70}$$

We need to relate this isotopes production to the burn up we defined earlier. This will allow us to use a similar methodology.

$$\frac{dN_{Pu-242}}{dt} = \frac{\sigma_{\gamma}^{Pu-241}}{\sigma_{f}^{Pu-239}} \frac{N_{Pu-241}(t)}{N_{Pu-239}(t)} \sigma_{f}^{Pu-239} N_{Pu-239}(t)\varphi(t) \tag{71}$$

$$\int_0^T dN_{Pu-242} = \frac{\sigma_{\gamma}^{Pu-241}}{\sigma_{f}^{Pu-239}} \frac{N_{Pu-241}(t)}{N_{Pu-239}(t)} \int_0^T \Sigma_{f}^{Pu-239}(t)\varphi(t)dt \tag{72}$$

$$\int_0^T dN_{Pu-242} = \frac{\sigma_{\gamma}^{Pu-241}}{\sigma_{f}^{Pu-239}} \frac{N_{Pu-240}(t)}{N_{Pu-239}(t)} \Psi \frac{1}{E_r} \rho^{Pu} \tag{73}$$

We will assume that the Pu-241 to Pu-239 ratio does not change significantly over time. Thus we are able to use the value calculated before in that place. This should under predict the value of the Pu-242 ratio since it does not take into consideration the production of Pu-241 which could become Pu-242. We will also substitute our definition of burn up to get:

$$N_{Pu-242}(T) - N_{Pu-242}(0) = \frac{\sigma_\gamma^{Pu-241}}{\sigma_f^{Pu-239}} \frac{N_{Pu-241}(0)}{N_{Pu-239}(T)} \Psi \frac{1}{E_r} \rho^{Pu} \qquad (74)$$

Then we will multiply by a constant in order to find the consistent factor for the burn up.

$$\frac{N_{Pu-242}(T) - N_{Pu-242}(0)}{N_{Pu-239}(T)} = \frac{\sigma_\gamma^{Pu-241}}{\sigma_f^{Pu-239}} \frac{N_{Pu-242}(0)}{N_{Pu-239}(T)} \Psi \frac{1}{E_r} \frac{\rho^{Pu}}{N_{Pu-239}(T)} \qquad (75)$$

Then with a little algebra we are able to find the following equation.

$$\frac{N_{Pu-242}(0)}{N_{Pu-239}(T)} = \frac{N_{Pu-242}(T)}{N_{Pu-239}(T)} - \frac{\sigma_\gamma^{Pu-241}}{\sigma_f^{Pu-239}} \frac{N_{Pu-241}(0)}{N_{Pu-239}(T)} \frac{1}{E_r} \frac{m_{Pu}}{N_a} \left\{ \Psi \frac{N_{Pu-239}(0)}{N_{Pu-239}(T)} \frac{N_{Pu}(0)}{N_{Pu-239}(0)} \right\} \qquad (76)$$

$$\frac{N_{Pu-242}(0)}{N_{Pu-239}(T)} = \frac{N_{Pu-242}(T)}{N_{Pu-239}(T)} - \frac{\sigma_\gamma^{Pu-241}}{\sigma_f^{Pu-239}} \frac{N_{Pu-241}(0)}{N_{Pu-239}(T)} \frac{1}{E_r} \frac{m_{Pu}}{N_a} \Psi_X \qquad (77)$$

Americium-241:

The derivation for Americium ratio is a little different than the plutonium isotopes. The key factor causing this difference is that each of the previous isotopes was being produced and not being lost. Since ^{241}Am is produced by the beta decay of ^{241}Pu. Over the time period we are going to be considering, the decay of ^{241}Pu is basically non-existent. Thus it will be neglected. We start again with our standard change rate equation:

$$\frac{dN_{Am-241}}{dt} = \{production\} - \{loss\} \tag{78}$$

$$\frac{dN_{Am-241}}{dt} = 0 - N_{Am-241}(t)\sigma_a^{Am-241}\varphi(t) \tag{79}$$

The loss mechanisms for ^{241}Am have been are the loss through neutron absorption. Either neutron induced fission or radiative capture is possible for ^{241}Am. Now we can go ahead with solving our differential equation:

$$\frac{dN_{Am-241}}{N_{Am-241}(t)} = -\sigma_a^{Am-241}\varphi(t)\frac{N_{Pu-239}(t)}{N_{Pu-239}(t)}\frac{\sigma_f^{Pu-239}}{\sigma_f^{Pu-239}}dt \tag{80}$$

$$\frac{dN_{Am-241}}{N_{Am-241}(t)} \tag{81}$$

$$= -\frac{\sigma_a^{Am-241}}{\sigma_f^{Pu-239}}\frac{1}{N_{Pu-239}(t)}N_{Pu-239}(t)\sigma_f^{Pu-239}\varphi(t)dt$$

$$\int_0^T \frac{dN_{Am-241}}{N_{Am-241}(t)} \tag{82}$$

$$= -\frac{\sigma_a^{Am-241}}{\sigma_f^{Pu-239}} \frac{1}{N_{Pu-239}(t)} \int_0^T N_{Pu-239}(t)\sigma_f^{Pu-239}\varphi(t)dt$$

$$ln[N_{Am-241}(t)]_0^T = -\frac{\sigma_a^{Am-241}}{\sigma_f^{Pu-239}} \frac{1}{N_{Pu-239}} \Psi \frac{\rho^{Pu}}{E_r} \tag{83}$$

Now we will need to simplify by exponentiation and simplifying the term in the exponential:

$$[N_{Am-241}(t)]_0^T = exp\left[-\frac{\sigma_a^{Am-241}}{\sigma_f^{Pu-239}} \frac{1}{N_{Pu-239}} \Psi \frac{\rho^{Pu}}{E_r}\right] \tag{84}$$

$$[N_{Am-241}(t)]_0^T = exp\left[-\frac{\sigma_a^{Am-241}}{\sigma_f^{Pu-239}} \Psi_X \frac{m^{Pu}}{N_a E_r}\right] \tag{85}$$

Since the exponent will give us a factor by which the ratio is increased or decreased, we find the relative change of the two points of time we are interested in.

$$\frac{N_{Am-241}(T)}{N_{Pu-239}(T)} = \frac{N_{Am-241}(0)}{N_{Pu-239}(T)} exp\left[-\frac{\sigma_a^{Am-241}}{\sigma_f^{Pu-239}} \Psi_X \frac{m^{Pu}}{N_a E_r}\right] \tag{86}$$

$$\frac{N_{Am-241}(0)}{N_{Pu-239}(T)} = \frac{N_{Am-241}(T)}{N_{Pu-239}(T)} exp\left[\frac{\sigma_a^{Am-241}}{\sigma_f^{Pu-239}} \Psi_X \frac{m^{Pu}}{N_a E_r}\right] \tag{87}$$

Summary of Derived Equations:

Decay Time:

$$T_{decay} = \frac{1}{\lambda_{Sr-89}} ln\left[\frac{N_{Y-89}(t)}{N_{Sr-89}(t)} + 1\right]$$

Strontium-89:

$$\frac{N_{Sr-89}(T)}{N_{Pu-239}(T)} = Y_{Sr-89}\frac{1}{E_r}\frac{m_{Pu}}{N_a}\Psi_X$$

Strontium-90:

$$\frac{N_{Sr-90}(T)}{N_{Pu-239}(T)} = Y_{Sr-90}\frac{1}{E_r}\frac{m_{Pu}}{N_a}\Psi_X$$

Zirconium-95:

$$\frac{N_{Zr-95}(T)}{N_{Pu-239}(T)} = Y_{Zr-95}\frac{1}{E_r}\frac{m_{Pu}}{N_a}\Psi_X$$

Plutonium-238:

$$\frac{N_{Pu-238}(0)}{N_{Pu-239}(T)} = \frac{N_{Pu-238}(T)}{N_{Pu-239}(T)} - \frac{\sigma_{n,2n}^{Pu-239}}{\sigma_f^{Pu-239}}\frac{1}{E_r}\frac{m_{Pu}}{N_a}\Psi_X$$

Plutonium-239:

$$\frac{N_{Pu-239}(0)}{N_{Pu-239}(T)} = 1 + \frac{\sigma_a^{Pu-239}}{\sigma_f^{Pu-239}} \frac{1}{E_r} \frac{m_{Pu}}{N_a} \Psi_X$$

Plutonium-240:

$$\frac{N_{Pu-240}(0)}{N_{Pu-239}(T)} = \frac{N_{Pu-240}(T)}{N_{Pu-239}(T)} - \frac{\sigma_\gamma^{Pu-239}}{\sigma_f^{Pu-239}} \frac{m_{Pu}}{N_a} \frac{1}{E_r} \Psi_X$$

Plutonium-241:

$$\frac{N_{Pu-241}(0)}{N_{Pu-239}(T)} = \frac{N_{Pu-241}(T)}{N_{Pu-239}(T)} - \frac{\sigma_\gamma^{Pu-240}}{\sigma_f^{Pu-239}} \frac{N_{Pu-240}(0)}{N_{Pu-239}(T)} \frac{1}{E_r} \frac{m_{Pu}}{N_a} \Psi_X$$

Plutonium-242:

$$\frac{N_{Pu-242}(0)}{N_{Pu-239}(T)} = \frac{N_{Pu-242}(T)}{N_{Pu-239}(T)} - \frac{N_{Pu-241}(0)}{N_{Pu-239}(T)} \frac{\sigma_\gamma^{Pu-241}}{\sigma_f^{Pu-239}} \frac{m_{Pu}}{N_a} \frac{1}{E_r} \Psi_X$$

Americium-241:

$$\frac{N_{Am-241}(0)}{N_{Pu-239}(T)} = \frac{N_{Am-241}(T)}{N_{Pu-239}(T)} exp\left[\frac{\sigma_{absorb}^{Am-241}}{\sigma_f^{Pu-239}} \frac{m_{Pu}}{N_a} \frac{1}{E_r} \Psi_X\right]$$

APPENDIX C

ORTHOGONAL RANDOM SAMPLING DATA CHART

	Number of Perturbed Variables											
Variable	One	Two	Three	Four	Five	Six	Seven	Eight	Nine	Ten	Eleven	Twelve
Mass of Pu	236.74	235.52	236.71	239.41	237.91	240.47	235.52	240.59	236.42	239.49	242.95	240.68
σ Pu-239 (n,f)	1.77E+00	1.74E+00	1.65E+00	1.77E+00	1.87E+00	1.77E+00	1.72E+00	1.94E+00	1.78E+00	1.81E+00	1.96E+00	1.76E+00
σ Pu-239 (n,γ)	3.86E-01	3.86E-01	3.97E-01	4.53E-01	4.03E-01	4.11E-01	3.43E-01	4.10E-01	3.85E-01	4.39E-01	4.60E-01	3.83E-01
σ Pu-239 (n,a)	2.16E+00	2.16E+00	2.16E+00	2.28E+00	2.13E+00	2.19E+00	2.15E+00	2.44E+00	2.16E+00	2.01E+00	2.32E+00	2.26E+00
σ Pu-239 (n,2n)	8.64E-04	8.64E-04	8.64E-04	8.64E-04	6.70E-04	5.73E-04	8.21E-04	8.08E-04	8.71E-04	5.16E-04	8.32E-04	7.02E-04
σ Pu-240 (n,γ)	4.46E-01	4.46E-01	4.46E-01	4.46E-01	4.46E-01	5.33E-01	5.18E-01	4.39E-01	1.90E-01	4.66E-01	4.22E-01	6.16E-01
σ Pu-241 (n,γ)	3.75E-01	3.75E-01	3.75E-01	3.75E-01	3.75E-01	3.75E-01	1.99E-01	4.59E-01	4.41E-01	4.13E-01	2.22E-01	3.41E-01
σ Pu-241 (n,a)	2.63E+00	2.63E+00	2.63E+00	2.63E+00	2.63E+00	2.63E+00	2.63E+00	2.45E+00	2.71E+00	2.74E+00	2.47E+00	2.81E+00
Y Sr-89	1.81E-02	1.81E-02	1.81E-02	1.81E-02	1.81E-02	1.81E-02	1.81E-02	1.81E-02	1.87E-02	1.80E-02	1.89E-02	1.70E-02
Y Sr-90	2.13E-02	2.13E-02	2.13E-02	2.13E-02	2.13E-02	2.13E-02	2.13E-02	2.13E-02	2.13E-02	2.08E-02	2.11E-02	2.01E-02
Y Zr-95	4.77E-02	4.77E-02	4.77E-02	4.77E-02	4.77E-02	4.77E-02	4.77E-02	4.77E-02	4.77E-02	4.77E-02	5.24E-02	4.61E-02
E_r	207	207	207	207	207	207	207	207	207	207	207	206.793
Results:												
Burn Up	5997.37	6027.72	5998.16	5929.17	5970.49	5904.34	6026.65	5899.54	5939.86	5987.49	5603.33	6198.78
Percent Error	0.92	0.42	0.91	2.05	1.37	2.46	0.44	2.54	1.87	1.09	7.43	-2.40
Pu-240/ Pu-239	0.050133	0.0500984	0.0500904	0.0498657	0.0501677	0.0500342	0.0502411	0.0501563	0.0501646	0.0499912	0.0501122	0.0500177
Percent Error	0.38	0.45	0.47	0.91	0.31	0.58	0.17	0.34	0.32	0.67	0.43	0.61

VITA

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